NANOFLUIDS THERMAL CONDUCTIVITY PREDICTIONS: A COMPARISON OF THEORETICAL APPROACHES

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Abstract. A nanofluid consist of an emulsion of fluid and dispersed particles of nanoscale size. Normally, the base fluid and the dispersed particles have quite different properties and the result is the considerable improvement of the thermophysical properties compared with base fluid, which in turn, allows the application of this emulsions for severe conditions into electronic circuits and sophisticated heat transfer equipments. Classical theoretical models have shown their limitations to accurately predict the nanofluids conductivities, specially, for volume fractions above 5% of particles volume and neglects thermal and particle size effects. Two approaches are presented aiming at explaining the anomalous behavior of the thermal conductivity of these systems: The fist one, termed Unit Cell Model (UCM) considers the Brownian movement experienced by the nanoparticles while the second one so-called Complex Nanoparticle Model(CNM) assumes a stationary interface between the base fluid and the particles. In this paper, these two approaches are analyzed and compared with experimental data. These two methodologies are applied to systems: TiO₂-H₂O and Al₂O₃-H₂O and discussed in light of the models limitations and volume fraction ranged from 0.5 to 5 % of particle volume. The effect of important parameters such as temperature, size diameter and geometrical model parameters were investigated suggesting improvements and refinement of these models.

Keywords: Nanofluids, theoretical models, Thermal conductivity

1. INTRODUCTION

A Nanofluid is a system constituted by mixture of solid particles of nanometrical size dispersed in a liquid. As the thermal physical properties of the particle usually have values different from the base fluids, these properties in the mixture are enhanced. Among these properties, the thermal conductivity, k, has special interest, not only due to discrepancies of value found in experimental works, but its relevance in applications involving processes of heat transfer, favoring the thermal changes (Eastman et al, 2004). Historically, aiming at increasing thermal performance, the addition of solid particles in base fluids has been studied, however, until approximately ten passed years, with rare exceptions, the applications have limited to milli-sized [mm] or micro-sized particles due to the difficulty on obtaining nano-size particles economically. The applications of milli and micro-sized particles dispersed on fluids encounters severe limitations because of sedimentation effects, obstruction of channels and abrasive effects (Lee et al, 1999). The prediction of the thermal conductivity of the resulting mixture, keff, can be outstanding the classic models of Bruggeman (Bruggman, 1935) and Hamilton-Crosser (Hamilton and Crosser, 1962), where they use in their formulations the thermal conductivity of the particle, k_p , the thermal conductivity of the base fluid, k_m and the volumetric fraction, v, that is defined as the reason between the total volume of the particles and the total volume of the mixture. In an innovative work, Choi (Choi, 1995) presents the idea of addition of particles of nanometrical size in a base fluid, calling nanofluid. The addition of nanoparticles in fluid base has shown potential application and high enhancement of thermal conductivity has been observed. The development of techniques and methodology for both produce nanofluids and use of nanoparticles in heat transfer equipments still is a challenge due to the dynamics of the particle in the emulsion, interactions and surface effects. Therefore, the motivation for new studies is growing and is necessary a step forward to reach industrial application in compacts heat transfer, in spit of relative success in small systems. The classical models to predict the enhancement of thermal conductivity of nanofluids has fail because they neglects important effects of nanoscale such as size diameters, interactions and temperature effects. Therefore, these models, when applied to nanofluid systems, do not capture in a perfected way the increments of the thermal conductivity of the mixture, motivating the study of new models (Eastman et al, 2004). Lee et al (Lee et al, 1999) and Wang et al (Wang et al, 1999) carried out the first experiment on nanofluids and obtained the results for the heat conductivity enhancement. Das et al (Das et al, 2003), study the increments of the thermal conductivity of the nanofluid as function of the temperature of the mixture, concluding that the thermal conductivity increase as temperature was increased. Xuan et al (Xuan et al, 2003), analyze the effects of the Brownian movement of the nanoparticle in nanofluid systems, detaching the relevance of this effect in the increments of the properties. Chom and Kihm (Chom and Kihm, 2005), study the influence of the size of the nanoparticle in the base fluid in the mixture results and Hong et al (Hong et al, 2005) and Murshed et al (Murshed et al, 2005) demonstrated the non-linear relationship to the increments of the thermal conductivity of the nanofluid in relation to the base fluid, k_{eff}/k_m , and the volume fraction, v. Recently, in the calculation of the increments of the thermal conductivity of the nanofluids, two approaches stand out. The first assumes thermal physic properties different in an interfacial layer between the nanoparticle and the base fluid. Based on this approach, Xue and Xu (Xue and Xu, 2005), create the "Complex Nanoparticle Model", CNM, inserting in the formulations additional considerations on the size of the nanoparticle and the interfacial properties. The second approach turns on the influence of the effects of the Brownian movement of the nanoparticle for the results in nanofluid systems. Hrishikesh et al (Hrishikesh et al, 2007), use this approach to generate "Unitary Cell Model", UCM, that include in the formulations additional considerations on the size of the nanoparticle and the nanofluid temperature. In this investigation, these two models are analyzed and applied for different systems. The model parameters are discussed and sensibility analysis is carried out in order to verify the relative contributions of model parameters on the thermal conductivity enhancement predicted by these models. Limitations of the models are discussed in light of phenomenological aspects of the real system.

2. METHODOLOGY

2.1. Complex Nanoparticle Model (CNM)

This model assumes that the interface region between the nanoparticle and the base fluid has significant influence for the results in the increment of the thermal conductivity of nanofluid. It is based on the definition of Complex Nanoparticle, that is the juxtaposition of the spherical nanoparticle of thermal conductivity, k_p , with radii R and a spherical shell, interfacial layer, of thermal conductivity k_i and thickness t, as show in Fig.1. This physical system is used to formulated the equations for predicting the increment of the thermal conductivity of the nanofluid in CNM. The formulation use, as base, the classic method of Bruggeman (Bruggeman, 1935), where the volume fraction is replaced by the volume fraction of the complex nanoparticle, v_c and the thermal conductivity of the solid substituted by effective conductivity of complex nanoparticle, k_c . Then the energy equations is solved in the domains of nanoparticle and fluid base. Following the principles of the model is addressed with appropriated boundary conditions.





Let the intensity vector, H, and the heat flux vector, q, defined for equation of Fourier, where ϕ is the temperature distribution function and k, the thermal conductivity.

$$\boldsymbol{q} = \boldsymbol{k} \boldsymbol{H} \tag{1}$$

$$H = -\nabla\phi \tag{2}$$

When a intensity H_0 is applied on the nanofluid system in the direction defined by the coordinate z, according to Fig. 1, without considering internal energy sources and steady state, the governing equation is the equation of Laplace, that written in spherical coordinates and rotational symmetry is given by:

$$\frac{\partial}{\partial r} \left(r^2 \frac{\partial \phi}{\partial r} \right) + \frac{1}{\operatorname{sen} \theta} \frac{\partial}{\partial \theta} \left(\operatorname{sen} \theta \frac{\partial \phi}{\partial \theta} \right) = 0 \tag{3}$$

The boundary conditions for this problem that establish the compatibilities of temperature distribution function, ϕ , and heat flux vector, q, in the region-limits nanoparticle/interface and interface/outside the complex nanoparticle, where ϕ_p , ϕ_i and ϕ_m are the temperature distribution functions in the nanoparticle, interfacial layer and outside the Complex nanoparticle, respectively. Also, k_p , k_i and k_m are the thermal conductivities of the nanoparticle, interfacial layer and base fluid, respectively.

(4)

 $\phi_p(r=0) = \text{constant}$

$$\phi_m(r \to +\infty) = -H_0 r \cos\theta \tag{5}$$

$$\phi_n(r=R) = \phi_i(r=R) \tag{6}$$

$$\phi_i(r=R+t) = \phi_m(r=R+t) \tag{7}$$

$$k_{p} \frac{\partial \phi_{p}}{\partial r} (r = R) = k_{i} \frac{\partial \phi_{i}}{\partial r} (r = R)$$
(8)

$$k_i \frac{\partial \phi_i}{\partial r} (r = R + t) = k_m \frac{\partial \phi_m}{\partial r} (r = R + t)$$
⁽⁹⁾

In the application of this equation to each defined sub-region of physical problem, nanoparticle, interfacial layer and outside the complex nanoparticle, with the respective boundary conditions, the solution is:

$$\phi_{p}(r,\theta) = \frac{-9k_{m}k_{i}H_{0}r\cos\theta}{(2k_{m}+k_{i})(k_{p}+2k_{i})-2\left(\frac{R}{R+t}\right)^{3}(k_{i}-k_{m})(k_{i}-k_{p})} ; r < R$$
(10)

$$\phi_{i}(r,\theta) = \frac{-3k_{m} \left[\left(2k_{i} + k_{p} \right)r + \left(2k_{i} - k_{p} \right)R^{3}r^{-2} \right] H_{0} \cos \theta}{\left(2k_{m} + k_{i} \right) \left(k_{p} + 2k_{i} \right) - 2 \left(\frac{R}{R+t} \right)^{3} \left(k_{i} - k_{m} \right) \left(k_{i} - k_{p} \right)} ; R < r < R+t$$
(11)

The thermal conductivity of the complex nanoparticle, k_c , is calculated by the Eq. (1) considering the heat flux vector, q, and the intensity vector, H, as space averages of the vectors q and H of the levels of nanoparticle and interface, respectively. Then, after algebraic manipulations:

$$k_{c} = k_{i} \frac{2k_{i} + k_{p} + 2\lambda(k_{p} - k_{i})}{2k_{i} + k_{p} - \lambda(k_{p} - k_{i})}$$
(12)

The volume fraction calculated with the complex nanoparticle, v_c is obtained, with base in the Fig. 1, through the geometric relationship:

$$v_c = \frac{v}{\left(\frac{R}{R+t}\right)^3} \tag{13}$$

The thermal conductivity of the complex nanoparticle, k_c , and the volume fraction calculated with the complex nanoparticle, v_c , are used in the equation of the classic model of Bruggeman (Bruggeman, 1935), presented to proceed. Equation (14) with the Eq. (13) and Eq. (12) define CNM, where the k_i and t values are estimated (Xue and Xu, 2005).

$$\frac{k_{eff}}{k_m} = \frac{(3v_c - 1)\frac{k_c}{k_m} + (2 - 3v_c) + \sqrt{\left((3v_c - 1)\frac{k_c}{k_m} + (2 - 3v_c)\right)^2 + 8\frac{k_c}{k_m}}}{4}$$
(14)

2.2. Unitary Cell Model (UCM)

This model considers the increase of the value of the thermal conductivity of the nanofluid, be due, mainly, to the micro-convection heat transfer, motivated by the Brownian movement of the nanoparticle. In UCM, the nanoparticles are considered as spherical, mono-dispersed and without gatherings with homogeneous distribution in the base fluid. The nanoparticles interaction is neglected. Based on these hypotheses, UCM has the limitation of being applied to low

volume fractions (Hrishikesh et al, 2007). The Fig. 2a defines the Unitary Cell, which serves for base for the development of the model. The UCM is one-dimensional, where the thermal conductivity of nanofluid is estimated by the combination of the mechanisms of conduction heat transfer in the nanoparticle, micro-convection in the interface base fluid/nanoparticle region due to the nanoparticles Brownian movement and conduction in the remaining of the base fluid, according to outline of the Fig. 2b, also showing the thermal resistances of each one of those effects.



Figure 2. (a) Definition of unitary cell and (b) thermal resistances in unitary cell for UCM application

To evaluate the increment of the thermal conductivity of the nanofluid in relation to the base fluid, k_{eff}/k_m , the heat transfers through the unitary cell are used with the nanoparticle, Q_{eff} , and without the nanoparticle, Q_m .

$$Q_{eff} = -k_{eff} A_m \frac{dT}{dx} = \frac{1}{R_{eff}} \Delta T$$
⁽¹⁵⁾

$$Q_m = -k_m A_m \frac{dT}{dx} = \frac{1}{R_m} \Delta T \tag{16}$$

$$\frac{k_{eff}}{k_m} = \frac{\frac{1}{R_{eff}}}{\frac{1}{R_m}}$$
(17)

With base in the outline of the Fig. 2b, the equivalent resistance, R_{eff}, is calculated by:

$$\frac{1}{R_{eff}} = \frac{1}{R_m} + \frac{1}{R_p + R_c}$$
(18)

After the calculations using the relative areas for heat transfers:

$$\frac{k_{eff}}{k_m} = 1 + \frac{k_p}{k_m} \left(\frac{L}{6d_p + \frac{k_p}{h}} \right) \frac{\pi d_p^2}{L^2}$$
(19)

For the obtaining of the convective heat transfer coefficient, h, is necessary the analysis of the contribution of the Brownian movement. With a local analysis (White, 1991) of the Nusselt and Peclet numbers, where C is an empiric coefficient, non-dimensional, function of peculiar characteristics of the nanoparticle / base fluid combination (Hrishikesh et al, 2007):

$$h = C \frac{u_p k_p}{\alpha_m} \tag{20}$$

In this equation, α_m is the thermal difusivity of the base fluid and u_p , the nanoparticle velocity in the Brownian movement, given by Keblinski et al (Keblinski et al, 2002), where k_b is the constant of Boltzmann, $k_b = 1,3807 (10)^{-23}$ J/K, and μ_m , the dynamic viscosity of the base fluid.

$$u_p = \frac{2k_b T}{\pi \mu_m d_p^2} \tag{21}$$

After the simplifications, the equation of UCM is given by:

$$\frac{k_{eff}}{k_m} = 1 + \frac{k_p}{k_m} \left(\frac{\pi}{6 + \frac{\alpha_m \,\mu_m \,\pi \,d_p}{2 \,C \,k_b \,T}} \right) \left(\frac{6 \,\nu}{\pi} \right)^{\frac{1}{3}} \tag{22}$$

3. APPLICATIONS

In each graph result of the applications, experimental data, curves generated by CNM (Xue and Xu, 2005), Eq. (14), and UCM (Hrishikesh et al, 2007), Eq. (22), and the classic method of Hamilton-Crosser (Hamilton and Crosser, 1962), MHC, are presented for effect of comparisons and conclusions. The parameters for CNM are estimated (Xue and Xu, 2005) in thermal conductivity of the interface, $k_i = 5W/(mK)$, and interface thickness, t = 3nm. For analysis of the global behavior of the application for each method the error norm, N, is defined by the Eq. (23), where j is the number of points in the application and e_i , the relative error of each considered point.

$$N = \sqrt{\frac{\sum_{i=1}^{j} e_i^2}{j}}$$
(23)

In the Fig. 3a is presented the application to a nanofluid system $TiO_2 - H_2O$ of increment of the thermal conductivity of the nanofluid in relation to the base fluid, k_{eff}/k_m , function of the volume fraction, v. The experimental data were obtained by Murshed et al (Murshed et al, 2005) for nanoparticle diameter, dp = 15nm and nanofluid temperature, T = 25°C. For UCM, C is used equal 7. The analysis of the data display that UCM, N = 3,8%, captures in a satisfactory way the data of the application, with exception for the smallest volume fractions. CNM, N = 6,7%, it generates a curve with format that doesn't accompany the tendency of the evolution of the experimental data. HCM, N = 12,1%, it presents values below the experimental ones, as it is the expected for a classic method, when they are used to analyze a nanofluid



Figure 3. Applications for increment of the thermal conductivity of the nanofluid in relation to the base fluid, $k_{eff}/k_{m.}$ (a) TiO₂ - H₂O system: function of volume fraction. (b) Al₂O₃ - H₂O system: function of nanofluid temperature.

(Eastman et al, 1999). In the Fig. 3b is presented the application to a nanofluid system $Al_2O_3 - H_2O$ of increment of the thermal conductivity, k_{eff}/k_m, function of the nanofluid temperature, T. The experimental data were obtained for Das et al (Das et al, 2003) for nanoparticle diameter, dp = 38,4nm and volume fraction, v = 0,04. For UCM, C from 2 to 3,5 is used varying lineally with absolute temperature. The analysis of the data display that UCM, N = 0.6%, captures in an appropriate way the data of the application, being the only of the methods considered sensitive for the calculations with the variation of the temperature. The linked explanation to this method is that, as larger the considered temperature, more active the Brownian movement, Eq. (21) and, consequently, larger the efficiency of the transmission of heat in the micro-convection, Eq. (20). HCM, N = 5.6%, and CNM, N = 5.6%, are not appropriate for this application, in spite of, in the calculations, the variations with the temperature of nanoparticle and base fluid thermal conductivities have been considered. In the figure 4 is presented the application to a nanofluid system $Al_2O_3 - H_2O$ for increment of the nanofluid thermal conductivity, k_{eff}/k_m , function of the nanoparticle diameter, dp. The experimental data were obtained by Chom et al (Chom et al, 2005) for nanofluid temperature, $T = 25^{\circ}C$ and volume fraction, v = 0,01. For MCU, C is used equal 2. The analysis of the data display that UCM, N = 1,3%, captures in an appropriate way the data for the application. CNM, N = 5.9%, generates curve format waited for the case, with values, however, of inferior quality to UCM. MHC, N = 9.7%, doesn't foresee in formulation differences of results function of the nanoparticle size. Before that, it is ended that is not adapted for this application type. The behavior of the increase of the thermal conductivity with the decrease of the size of the particle, for a certain volume fraction, it is waited, since, staying the total volume of the particles and diminishing the size of these, does with that, consequently, a larger number of particles be had and then, a larger area of change of heat (Hrishikesh et al, 2007).



Figure 4. $Al_2O_3 - H_2O$ system: applications for increment of the thermal conductivity of the nanofluid in relation to the base fluid, k_{eff}/k_m , function of nanoparticle diameter.

4. ANALYSIS OF THE MODELS

In relation to CNM, Eq. (14), the increment of the thermal conductivity, k_{eff}/k_m , is function of the thermal conductivities of the nanoparticle, k_p , of the base fluid, k_m , and of the interface, k_i ; of the radii of the particle, R; of the volume fraction, v, and of the interface thickness, t. For a given k_p , k_m , R and v conditions, the results for the improvement of the thermal efficiency of the nanofluid are determined by the appropriate estimate of t and k_i . However,



Figure 5. TiO₂ - H₂O system (Fig 3a) – CNM analysis: error norm for (a) t = 3nm and (b) $k_i = 5$ W/(mk)

taking for base the experimental data of the application of the Fig. 3a and, for inspection, to substitute values of t and k_i in the Eq. (14), the conclusion there is not possibility to obtain a result with quality. The Fig. 5 exemplify the fact where staying constant t or k_i, the error norms, N, are not reduced to smaller values than 6,3%. Based on this analysis, the results without quality obtained by CNM, therefore, are conditioned to the limitations of the own method and not of inadequate estimates of the variables k_i and t. MCU has as main advantage the simplicity. The one-dimensional hypothesis is the principal limit factor, since the problem is strictly three-dimensional. As consequence, the coefficient empiric non-dimensional, C, Eq. (20), function of intrinsic characteristics in the nanoparticle / base fluid relationship, has the responsibility of adjusting the results close to the experimental data. For the application of UCM, Hrishikesh et al (Hrishikesh et al, 2007) suggests that, for a system nanofluid constituted of an oxide and water, C assumes value of the order of 10° . The solution of the Eq. (22), however, generates results in a great strip for different values of C in this order of greatness. The Fig. 6a presents for the system TiO_2 - H_2O , multiple curves, generated for UCM, varying C, in the calculation of the increment of the conductivity thermal function of the volume fraction. The Fig. 6b presents the error norm, N, for the same application, where several values of C are used in the solution of the Eq. (22). It can be observed that values of C, inside of the guided strip, generate some solutions of little quality. For the global analysis, for the simplify hypotheses for the UCM generation, it is waited that the model has better acting in the strip where the volume fraction, v, is relatively small (Hrishikesh et al, 2007). The analysis of the system TiO₂ - H₂O, Fig. 3a, however, it shows the opposite. The reason of this discrepancy is in the assembly of the outline of thermal resistances, Fig. 3b. In the inferior line of the heat flow, is considered that the influence of the micro-convection extends in a characteristic length of value of difference beside the cell side and the nanoparticle diameter. In the consideration of small values for the volume fraction, the nanoparticle has tiny dimensions when compared with the own cell and, therefore, the influence of the micro-convection is limited to the proximities of the nanoparticle.



Figure 6. TiO_2 - H_2O system (Fig. 3a) –UCM analysis: (a) multiple curves varying C, in the calculation of the increment of the conductivity thermal function of the volume fraction; (b) error norm, where several values of C are used.

4. CONCLUSION

In the direct comparison of the methods, UCM takes advantage of CNM, be in the format of resulting curves as in the increments of conductivity thermal functions of the volume fraction, of the temperature or of the particle size. For CNM, there is the need of analysis of the terms k_i and t for the k_c definition. The use of the equation of Bruggeman as base for the development of CNM is debatable, since the format of the resulting curve commits the results. The coefficient C, in UCM, needs to still be explored so that its order of magnitude would be better estimated. An optimization in MCU, foreseeing limited performance for the micro-convection provoked by the Brownian movement should be implemented, mainly due to its poor predictions for volume fractions very small.

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