

## Model for Heat Conduction in Nanofluids

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A comprehensive model has been proposed to account for the large enhancement of thermal conductivity in nanofluids and its strong temperature dependence, which the classical Maxwellian theory has been unable to explain. The dependence of thermal conductivity on particle size, concentration, and temperature has been taken care of simultaneously in our treatment. While the geometrical effect of an increase in surface area with a decrease in particle size, rationalized using a stationary particle model, accounts for the conductivity enhancement, a moving particle model developed from the Stokes-Einstein formula explains the temperature effect. Predictions from the combined model agree with the experimentally observed values of conductivity enhancement of nanofluids.

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Nanofluids or liquids with suspended nanoparticles are likely to be the future heat transfer media as their thermal conductivities are significantly higher than those of the parent liquids even when the nanoparticle concentrations are negligible. None of the existing theories is capable of explaining this anomalous increase. In this Letter, a theoretical model is evolved in steps to highlight the effects of various factors such as particle radius, concentration, and the temperature of the medium on the extent of enhancement in thermal conductivity.

Starting with the pioneering work of Maxwell [1], several authors [2–6] have developed theories for the effective thermal conductivity  $k$  of multiphase systems. Adler [6] has dealt with systems such as solid dispersed in liquid. The enhancement achieved in a nanofluid [7,8] is far in excess of the levels predicted by the above multiphase models. Several efforts [9,10] have been made in recent times to theorize this phenomenon. Kebinski *et al.* [10] presented four possible mechanisms, such as including Brownian motion of particles, layering of liquid molecules at the particle-liquid interface, ballistic nature of heat transport in nanoparticles, and nanoparticle clustering, to account for this anomalous increase. While these possible mechanisms are able to explain the thermal behavior partially, a comprehensive model is yet to emerge.

The present authors and their co-workers [11,12] have carried out experiments on oxide and pure metallic particle suspensions and brought out the strong influence of temperature on the enhancement of thermal conductivity, which the above mechanisms fail to explain. In the case of metallic particles, enhancement up to 9% was observed even at vanishing concentrations such as 0.000 26% [12], over the same temperature range.

Yu and Choi [4] used a renovated Maxwell model, with adjustable parameters such as liquid layer thickness. Yu *et al.* [13] presented a collision based mechanistic model

but could only predict the order of magnitude of enhancement. Moreover, these models do not explain the strong dependence of enhancement on temperature and particle size. The only model claimed to incorporate the effect of temperature on the conductivity enhancement was by Xuan *et al.* [14]. However, the dependence suggested by them is too weak ( $\propto T^{1/2}$ ) and they did not present any evidence for it either. Also, subsequent experiments [11,12] did not agree to such a dependence. In this Letter, a stationary particle model, which explains the dependence of  $k$  on the particle size and volumetric concentration, is developed first. As the next step, the model is extended to include the motion of nanoparticles. Microconvective heat transfer by moving particles is taken into account in terms of the particle velocity, which itself is taken as equal to that of the Brownian motion. Such an assumption explains the strong dependence of enhancement on temperature.

It is well known that, in a condensed phase such as liquid, short-range order prevails between the liquid molecules. Hence the liquid medium itself, for the sake of the model, can be looked upon as one comprised of particles, which interact with the nanoparticles. Since heat transfer is a surface phenomenon, its magnitude will increase with an increase in surface area of all the particles. In the case of nanoparticles, the surface area to volume ratio is much higher compared to the microparticles. This large total surface area offered by nanoparticles explains the anomalous increase in heat transfer rate (or effective thermal conductivity), even when a small volume fraction of nanoparticles is introduced in the liquid medium.

The proposed model incorporates the following two aspects:

(i) *Stationary particle model*.—In this approach, it is assumed that there are two parallel paths of heat flow

through the suspension, one through the liquid particles and the other through the nanoparticles.

The overall heat transfer rate of the system,  $q$  for one-dimensional heat flow may be expressed as

$$q = q_m + q_p, \quad \therefore q = -k_m A_m \left( \frac{dT}{dx} \right)_m - k_p A_p \left( \frac{dT}{dx} \right)_p, \quad (1)$$

where  $A$ ,  $k$ , and  $(\frac{dT}{dx})$  denote the heat transfer area, the thermal conductivity, and the temperature gradient of the respective medium, while the subscripts  $m$  and  $p$  denote quantities corresponding to the liquid medium and the particle phase, respectively. Assuming that the liquid medium and the nanoparticles are in local thermal equilibrium at each location, we can set

$$\left( \frac{dT}{dx} \right)_m = \left( \frac{dT}{dx} \right)_p = \left( \frac{dT}{dx} \right). \quad (2)$$

Now the overall heat transfer rate can be expressed as

$$q = -k_m A_m \left( \frac{dT}{dx} \right) \left[ 1 + \left( \frac{k_p A_p}{k_m A_m} \right) \right]. \quad (3)$$

It is proposed that the ratio of heat transfer areas  $A_p/A_m$  could be taken in proportion to the total surface areas of the nanoparticles ( $S_p$ ) and the liquid species ( $S_m$ ) per unit volume of the suspension. Taking both the liquid and the suspended particles to be spheres of radii  $r_m$  and  $r_p$ , respectively, the total surface area can be calculated as the product of the number of particles  $n$  and the surface area of the particle for each constituent. Denoting the volume fraction of the nanoparticles as  $\varepsilon$  and the volume fraction of the liquid as  $(1 - \varepsilon)$ , the number of particles for the two constituents can be calculated as

$$n_m = \frac{1 - \varepsilon}{(4/3\pi r_m^3)} \quad \text{and} \quad n_p = \frac{\varepsilon}{(4/3\pi r_p^3)}. \quad (4)$$

The corresponding surface areas of the liquid medium and the nanoparticle phase are given by

$$S_m = n_m \cdot 4\pi r_m^2 = 3 \frac{(1 - \varepsilon)}{r_m} \quad \text{and} \quad S_p = n_p \cdot 4\pi r_p^2 = \frac{3\varepsilon}{r_p}. \quad (5)$$

Taking  $\frac{S_m}{S_p} = \frac{A_m}{A_p}$  and substituting from Eq. (5) into the expression for heat transfer rate in Eq. (3), we obtain

$$q = -k_m A_m \left( \frac{dT}{dx} \right) \left[ 1 + \frac{k_p \varepsilon r_m}{k_m (1 - \varepsilon) r_p} \right] = -k_{\text{eff}} A_m \left( \frac{dT}{dx} \right), \quad (6)$$

where the effective thermal conductivity is expressed as

$$k_{\text{eff}} = k_m \left[ 1 + \frac{k_p \varepsilon r_m}{k_m (1 - \varepsilon) r_p} \right]. \quad (7)$$

The rate of heat transfer can be expressed in dimension-

less form as

$$q^* = \frac{q}{-k_m A_m (\frac{dT}{dx})} = 1 + \frac{k_p \varepsilon r_m}{k_m (1 - \varepsilon) r_p} = \frac{k_{\text{eff}}}{k_m}, \quad (8)$$

where the numerator represents the rate of heat transfer of the nanoparticle suspension, and the denominator is the rate of heat transfer in the absence of nanoparticles. In the above expression, it is seen that the enhancement is directly proportional to the ratio of conductivities and volume fraction  $\varepsilon$  of the nanoparticles (for  $\varepsilon \ll 1$ ) and is inversely proportional to the radius of the nanoparticles. In our laboratory, experiments are being carried out to verify the dependence of enhancement on various factors. The linear dependence of thermal conductivity enhancement on particle concentration is shown in Fig. 1.

As for the particle size effect, the above model shows that for a given liquid medium, the enhancement in effective conductivity increases with the reciprocal of nanoparticle radius. Available data [15] and our studies of different systems seem to agree with the inverse dependence of enhancement with particle size as shown in Fig. 2.

*(ii) Moving particle model.*—Our earlier studies have shown a strong dependence of enhancement in effective conductivity with respect to changes in temperature of the nanofluids [11,12]. In the present model, this strong dependence on the temperature of the medium is attributed to the variation of the velocity of the nanoparticles with temperature. In fact, the effective thermal conductivity of the particle is directly proportional to its mean velocity according to the kinetic theory [16]. Thus, it is possible to express the thermal conductivity of

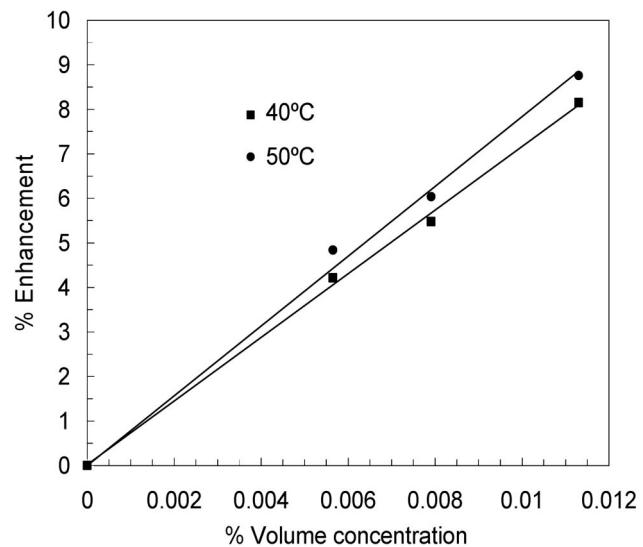


FIG. 1. Thermal conductivity enhancement with particle concentration for Au of 4 nm size in toluene.

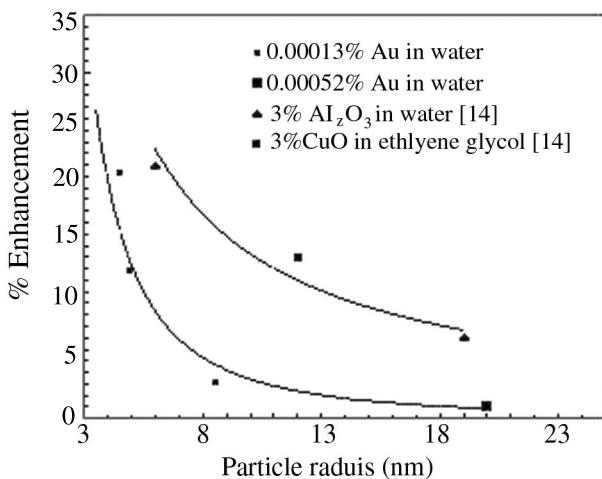


FIG. 2. Thermal conductivity enhancement with the nanoparticle size.

the particle as

$$k_p = c \cdot \bar{u}_p, \quad (9)$$

where  $\bar{u}_p$  is the average particle velocity and  $c$  is a constant. Combining Eqs. (7) and (9), we get the complete model, which is used to calculate the variation in effective thermal conductivity with temperature. The thermal conductivity enhancement taking into account the Brownian motion of particles can therefore be expressed as

$$\frac{k_{\text{eff}}}{k_m} - 1 = c \cdot \bar{u}_p \frac{\varepsilon r_m}{k_m(1 - \varepsilon)r_p}, \quad (10)$$

$$\therefore \text{enhancement of conductivity} \propto \bar{u}_p [\varepsilon/(1 - \varepsilon)] 1/r_p. \quad (11)$$

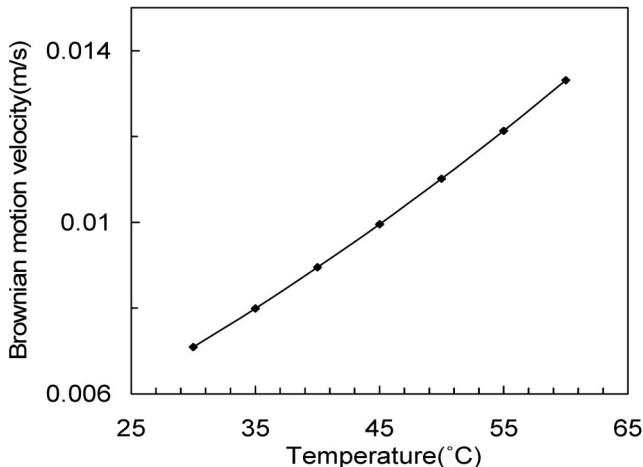


FIG. 3. Brownian motion velocities of Au particles of 17 nm mean diameter as a function of temperature.

It is proposed here that the velocity of the particles can be taken as that due to Brownian motion at a given temperature of the suspension. Mean velocity of the nanoparticle has been calculated using the Stokes-Einstein's formula,

$$\bar{u}_p = \frac{2k_b T}{(\pi \eta d_p^2)}. \quad (12)$$

It is evident from the above expression that the particle velocity ( $\bar{u}_p$ ) depends upon the factor  $T/\eta$ , where  $\eta$  is the dynamic viscosity of the fluid medium and  $T$  is the temperature. Since viscosity of the medium such as water decreases significantly with temperature, particle velocity increases as shown in Fig. 3. This explains the strong dependence of thermal conductivity enhancement with temperature.

The comparison between the predictions of the theoretical model given by Eq. (10) and our experimental data is shown in Fig. 4. A close comparison between the theoretically predicted and experimentally measured conductivity enhancement illustrates that the present model correctly explains the anomalous increase in effective thermal conductivity with temperature.

For the experimental conditions of the results presented in Fig. 4, the value of  $c$  is evaluated to lie between 2.9 and 3. An approximate estimate using kinetic theory shows that  $c \approx \frac{n l \hat{c}_v}{3}$ , where  $n$  is the particle concentration,  $l$  is the mean free path, and  $\hat{c}_v$  is the specific heat per particle. Using the Debye model for the particle,  $\hat{c}_v = 3Nk$ , where  $N$  is the number of atoms and  $k$  is the Boltzmann constant. For a typical particle having a diameter of 10 nm, the number of atoms per particle  $N \sim 10^6$ ,  $n \sim 10^{18}$ ,  $l \sim 10^{-2}$ , and  $\hat{c}_v \sim 10^{-16}$  J/K.

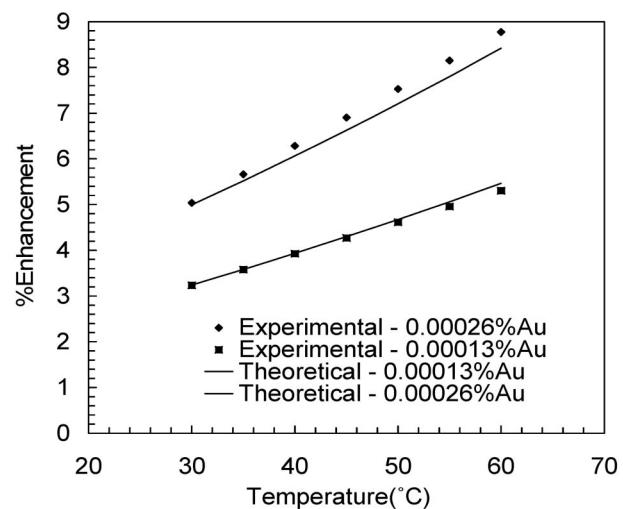


FIG. 4. Comparison of experimental and theoretical enhancement against temperature for a Au nanofluid of 17 nm mean diameter.

Using these estimates,  $c$  is found to be of the order of unity.

In summary, a comprehensive theoretical model has been developed which explains the enhancement in thermal conductivity of a nanofluid with respect to variation in particle size, particle volume fraction, and temperature. It is shown that in nanoparticle suspensions, the large enhancement of heat transfer rate (alternatively, enhancement in effective thermal conductivity) occurs due to the large surface area offered by the nanoparticles even at vanishing concentrations. This enhancement is inversely proportional to the radius of the particle since the surface area/volume ratio for the particle varies as  $1/r_p$ . The enhancement is linearly proportional to nanoparticle concentration, for small concentrations. The temperature dependence is attributed to the variation of Brownian motion velocity for the particles, which varies in proportion to  $T/\eta$ . Experimental results strongly support the predictions of the present model.

Even though the present model takes an important step towards understanding the heat conduction in nanofluids, its validity in the molecular size regime has to be explored. This model may not be used for a large concentration of the particles where interparticle interactions become important. A precise understanding of the constant  $c$  in Eq. (9) and the liquid particle size  $r_m$  may improve this model.

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