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Thermal diffusion in dilute nanofluids investigated by photothermal interferometry

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Abstract. We have carried out a theoretical analysis of the dependence of the particle mass fraction on the thermal diffusivity of dilute suspensions of nanoparticles in liquids (dilute nanofluids). The analysis takes in to account adsorption of an ordered layer of solvent molecules around the nanoparticles. It is found that thermal diffusivity decreases with mass fraction for sufficiently small particle sizes. Beyond a critical particle size thermal diffusivity begins to increase with mass fraction for the same system. The results have been verified experimentally by measuring the thermal diffusivity of dilute suspensions of TiO$_2$ nanoparticles dispersed in polyvinyl alcohol (PVA) medium. The effect is attributed to Kapitza resistance of thermal waves in the medium.

1. Introduction
During the past few years the thermal transport properties of fluids with low concentration suspensions of nanoparticles, the so called nanofluids, have puzzled the scientific community. Nanofluids have offered theoretical challenges because in many cases the measured thermal properties of nanofluids containing extremely low concentrations (much less than 0.1 volume %) of nanoparticles is often an order of magnitude larger than that predicted by existing models, and the experimental values reported by different groups differ significantly. In this work we have carried out a theoretical analysis and experimental verification of the variation of the normalized thermal diffusivity of a dilute nanofluid with nanoparticle concentration. The analysis takes in to account adsorption of the liquid around the nanoparticles forming a layer. The analysis is based on an extended Maxwell-Garnett model, which considers the particles as non-interacting and isolated in the host medium. We show that thermal diffusion in dilute nanofluids is possibly controlled by Kapitza resistance. We have confirmed the theoretical results by measuring thermal diffusivity of low concentrations of nano TiO$_2$ dispersed in Poly vinyl alcohol by thermal wave interferometry in a resonant cavity.

2. Theoretical background
We have carried out numerical computations of the normalized thermal diffusivity of Polyvinyl alcohol (PVA) and water based dilute nano suspensions, following the extended Maxwell – Garnett (M-G) model. The thermal conductivity of nanofluids, as predicted by M-G model has been modified including the effects of particle size and adsorption of liquid layer around the particle surface [1]. This approach postulates the formation of a semisolid-like layer of liquid around the nanoparticle surface, which has a relatively high thermal conductivity compared to the base liquid. Adsorption gives rise to
a modified volume fraction, which has dependences both on particle size and on the surrounding layer thickness. This fluid adsorption layer on the particle surface affects the thermal diffusivity of the medium very strongly.

For a two-component entity of spherical particle suspension, the M - G Model gives the following expression for the effective thermal conductivity \( k \) of the nanofluid [2].

\[
\frac{k}{k_f} = \frac{(1 - \phi_v)(k_p + 2k_f) + 3\phi k_p}{(1 - \phi_v)(k_p + 2k_f) + 3\phi k_f}
\]  

(1)

where, \( k_f \) and \( k_p \) are the thermal conductivities of the host fluid medium and the particle (in the bulk) respectively. \( \phi_v \) is the volume fraction of particles in the medium, which can be estimated from the corresponding mass fraction \( \phi_m \) as

\[
\phi_v = \frac{\phi_m \rho_f}{\rho_p + \phi_m \rho_f - \phi_m \rho_p}
\]  

(2)

Using the relaxation time approximation, the effective thermal conductivity of nonmetallic nanoparticles can be approximated as [3]

\[
k_p = \frac{3a^*}{4 + 1}k_{bp}
\]  

(3)

where \( k_{bp} \) is the bulk thermal conductivity of the particle, \( a^* = a/l \) is the non-dimensional radius and \( l \) is the mean free path of the phonons within the particles.

The expressions for the effective density and effective specific heat of nanofluids are given by [4]

\[
\rho_{eff} = \rho_p \phi_v + \rho_f (1 - \phi_v)
\]  

(4)

\[
C_{eff} = C_{pf} \phi_v + C_{pf} (1 - \phi_m)
\]  

(5)

The formation of solid-like layers leads to a larger effective volume fraction for the particles. With the consideration of the interface layer, one should substitute \( \phi_v \) in equations (1) to (5) by \( \phi_{rad} = [(t + a)/a]\phi_v \), which is the virtual volume fraction of the particles. After replacing \( k \), \( \rho \) and \( C_p \) by \( k_{eff} \), \( \rho_{eff} \) and \( C_{eff} \) respectively, the effective thermal diffusivity can now be estimated from the relation

\[
\alpha = \frac{k}{\rho C_p}
\]  

(6)

The evaluated values have been normalized with the thermal diffusivity of the base fluid. Theoretical results for dilute nanofluids of TiO\(_2\) in PVA and TiO\(_2\) in water are shown in figures 1 and 2. It can be seen that for dilute nanofluids with low particle sizes, the thermal diffusivity decreases with particle concentration, but the effect decreases as the particle size increases and vanishes at high enough sizes.

4. Experimental method

Nanofluids with mass fractions 0.005%, 0.01% and 0.039% were prepared by the normal dispersion technique. TiO\(_2\) nanoparticles of average size 15nm were first prepared by the hydrolysis of TiCl\(_4\) [5] and dispersed in the base fluid. 4% PVA and double distilled water were used as the base fluids. After dispersion the fluids were sonicated for about 30 minutes for stabilization. A Thermal Wave Resonant Cavity (TWRC) set up [6] was fabricated (shown in Fig. 3) to measure the thermal diffusivity of the fluid samples.
The resonant cavity was formed with a thin copper foil of thickness 37 µm on one side and a Ni-Cr metal coated PVDF film of thickness 28 µm on the other side. Cavity length as well as modulation frequency scans produced resonance like extrema in the lock-in amplifier in-phase and quadrature outputs, which could be used to determine the thermal diffusivity of the liquid, filled in the cavity. We have used the cavity scan technique in our measurements, since it provides accuracies better than 0.1%. Experimentally, two successive extrema in the in-phase signal amplitudes, $L_2(P)$ and $L_1(P)$, yielded necessary information required to compute thermal diffusivity of the medium. It was found that in-phase signals had better reproducibility than quadrature signals and so in-phase signals were used in our measurements. The thermal diffusivity of the fluid in the cavity was determined from the relation
\[ \alpha_s = \frac{f}{\pi}[L_2(P) - L_1(P)]^2 \]  

(7)

where \( f \) was the modulation frequency used in the experiment.

Figure 3: Block diagram of the TWRC set up

The TWRC set up had been tested with known fluids prior to making measurements in nanofluids.

5. Results and discussion

The measured variations of thermal diffusivity with particle concentration for dilute nanofluids of TiO$_2$ in PVA are shown in figure 4. It is seen that for small particle sizes the thermal diffusivity decreases as particle mass fraction increases. This observation is in agreement with the theoretical predictions based on the extended M-G model. The slope of the curve decreases with increase in particle size, and for a high enough particle size thermal diffusivity begins to increase with particle concentration. For small particle sizes in dilute concentrations a semisolid layer of the liquid is formed around each particle. The interfacial thermal resistance, or Kapitza resistance, caused by possible scattering of thermal waves at the boundary between the semisolid layer and the particle dominates in this regime. This Kapitza resistance gets less and less significant as grain sizes increase because the magnitude of the thermal conductivity of the material of the particles begins to play a larger role, and the parameters that appear in the effective medium theory begin to control the net thermal diffusivity of the mixture. Since the modified volume fraction due to adsorption and fluid layer formation around the particles depend on the nanoparticle size and adsorption layer thickness, the rate of decrease of thermal diffusivity with particle density (or mass fraction) will be higher for lower particle sizes. This will also vary with the density of the base fluid as the adsorption layer thickness strongly depends on the molecular weight of the fluid.
Figure 4: Normalized thermal diffusivity and thermal conductivity variation with mass fraction for dilute PVA based nanosuspensions of 15nm sized TiO$_2$ nanoparticles

6. Conclusions
The influence of nanoparticle sizes and mass fractions on the thermal diffusivity of dilute nanofluids have been systematically investigated and reported. It is demonstrated that thermal diffusivity measurements are more sensitive than thermal conductivity measurements to microscopic effects in systems such as nanofluids. More work to understand the effect of particle size, thermal conductivity of the material of the nanoparticle in the bulk form, influence of particle concentration on thermal diffusivity at higher concentrations etc. will throw more light on the mechanisms involved in this complex system.

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