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Formulation of nanofluids for natural convective heat transfer applications

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Abstract

The paper is concerned about formulation of aqueous based nanofluids and its application under natural convective heat transfer conditions. Titanium dioxide nanoparticles are dispersed in distilled water through electrostatic stabilization mechanisms and with the aid of a high shear mixing homogenizer. Nanofluids formulated in such a way are found very stable and are used to investigate their heat transfer behaviour under the natural convection conditions. The preliminary results are presented in this paper. Both transient and steady heat transfer coefficients are measured and the results show a systematic decrease in the natural convective heat transfer coefficient with increasing particle concentration. This is in contradiction to the initial expectation. Possible reasons for the observations are discussed.

Keywords: Nanofluids; Formulation; Natural convection; Electrostatic stabilization

1. Introduction

Recent advances in nanotechnology have allowed development of a new category of fluids termed *nanofluids*, which was first used by a group in Argonne National Laboratory USA (Choi, 1995) to describe liquid suspensions containing nanoparticles with thermal conductivities orders of magnitudes higher than the base liquids, and with sizes significantly smaller than ~100 nm. Many experimental studies have shown that nanofluids have much higher thermal conductivities than those predicted by macroscopic models (Masuda et al., 1993; Choi, 1995; Eastman et al., 1996; Eastman et al., 2001; Lee et al., 1999; Xuan and Li, 2000; Xie et al., 2002; Wang et al., 2003; Wen and Ding, 2004a; Hong et al., 2005). The augment of thermal conductivity could provide a basis for an enormous innovation for heat transfer intensification, which is pertinent to a number of industrial sectors including transportation, power generation, micro-manufacturing, chemical and metallurgical industries, as well as heating, cooling, ventilation and air-conditioning industry.

Past decade has seen increasing research activities in heat intensification using nanofluids. The published studies can be classified into three categories, namely, effective thermal conductivity under the static conditions, convective heat transfer (Pak and Cho, 1999; Xuan and Li, 2003; Wen and Ding, 2004b; Roy et al., 2004; Maiga et al., 2004; Yang et al., 2005) and phase change heat transfer (Das et al., 2003a,b; You et al., 2003; Tu et al., 2004; Wen and Ding, 2005). Most of the studies from the first category showed that nanofluids exhibited much higher thermal conductivities than those of base liquids (i.e., Masuda et al., 1993; Choi, 1995; Eastman et al., 1996; Lee et al., 1999; Xuan and Li, 2000; Xie et al., 2002; Wang et al., 2003; Wen and Ding, 2004a; Hong et al., 2005), and the enhancement increased with concentration and was much higher than predictions from macroscopic models. Many models have been proposed to explain such abnormal behaviours (Keblinski et al., 2002; Xie et al., 2002; Bhattacharya et al., 2004; Jang and Choi, 2004).

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However there are many inconsistent reports on nanofluids behaviour under other heat transfer conditions. *For forced convective heat transfer*, Pak and Cho (1999) found that the Nusselt number of Al_2O_3/water and TiO_2/water nanofluids increased with increasing volume fraction of suspended nanoparticles and Reynolds number. However, for a given average fluid velocity, the convective heat transfer coefficient for nanofluids with a volume fraction of 0.03 was found to be 12% lower than that of pure water. Similar observation has also been reported by Yang et al. (2005) for graphite nanofluids in laminar flow regime. The increase of experimental heat transfer coefficients was found to be lower than the enhancement of the effective thermal conductivity. Many other researchers, however, have reached opposite conclusions (i.e., Lee and Choi (1996); Xuan and Li, 2003; Wen and Ding, 2005). Lee and Choi (1996) reported heat transfer behaviour in parallel channels using an unspecified nanofluids and found a reduction in thermal resistance by a factor of 2. Xuan and Li (2003) studied Cu/water nanofluids heat transfer under turbulent flow conditions and showed substantial heat transfer enhancement. Wen and Ding (2004b) investigated heat transfer of Al_2O_3/H_2O nanofluids under laminar flow conditions and showed that presence of nanoparticles did enhance the convective heat transfer. The enhancement was shown to be much higher than the increase of the effective thermal conductivity, especially in the entrance region. *Controversial experimental results have also been observed for nanofluids heat transfer under pool boiling* conditions. Das et al. (2003) investigated nucleate pool boiling heat transfer of Al_2O_3/H_2O nanofluids using cylindrical cartridge heaters. The presence of nanoparticles was found to deteriorate boiling performance and the degradation was found to increase with nanoparticle concentrations. Deteriorated pool boiling heat transfer was also observed by Li et al. (2003) for CuO/H_2O nanofluids, which was thought to be the decreasing of active nucleation sites from nanoparticle sedimentation. To the other side, however, significant pool boiling heat transfer enhancement was observed for Al_2O_3/H_2O nanofluids (Tu et al., 2004) and for gold–water nanofluids (Witharana, 2003). For pool boiling at high heat fluxes, remarkable increase in the critical heat flux was also reported (You et al., 2003; Vassallo et al., 2004).

Compared with the research effort in forced convective and phase change heat transfer, relatively few studies have been carried on natural convective heat transfer. Khanafer et al. (2003) numerically investigated the heat transfer behaviour of nanofluids in a two-dimensional horizontal enclosure. The random motion of nanoparticles was considered through a dispersion model similar to thermal dispersion models for flows through porous media. The simulations showed that suspended nanoparticles substantially increased heat transfer at any given Grashof number. Such enhancement increased with particle concentration, which was thought to be the increased energy exchange from enhanced irregular and random movements of particles. However different experimental results have been observed by Putra et al. (2003) for natural convective heat transfer of aqueous CuO and Al_2O_3 nanofluids inside a horizontal cylinder. Unlike the results of thermal conduction and forced convection, experiments at Rayleigh number from 10^6 to 10^9 showed a systematic and significant deterioration in natural convective heat transfer. The deterioration increased with particle concentration and was more pronounced for CuO nanofluids. For example at a given Rayleigh number of $\sim 5 \times 10^7$, ~300% and ~150% decrease in the Nusselt number was observed for 4 wt% of CuO and Al_2O_3 nanofluids, respectively. The aspect ratio of the cylinder was also observed to influence heat transfer greatly.

The above short review shows that heat transfer behaviour of nanofluids is very complicated and application of nanofluids for heat transfer intensification should not be decided *only* by the effective thermal conductivity. Many other factors such as particle size, morphology and distribution of particles in the fluids could significantly influence the flow and heat transfer behaviour of nanofluids. This, however, has not been addressed properly so far. It is noted that the formulation of stable nanofluids still presents a big challenge, particularly for applications at elevated temperatures. Nanofluids prepared through dispersing dry nanoparticles generally have a stability problem (Xuan and Li, 2000; Das et al., 2003a; Wang et al., 2003). These nanofluids settle slowly after a period of time depending on the properties of nanoparticles, host liquids, surfactant/dispersant used, as well as the extent of agglomeration of nanoparticles. The instability of nanofluids could result in similar problems as encountered in micron or millimeter sized particulate dispersions, leading to sedimentation and even clogging of the system. This will no doubt deteriorate the heat transfer and could be one of the major reasons responsible for the observed difference in heat transfer behaviours. This work aims principally to formulate stable nanofluids that can be used under different conditions. Some experiments are also done on the use of the stable nanofluids under natural convective heat transfer conditions.

2. Experiment

2.1. Formulation and characterization of nanofluids

The properties and behaviour of a nanofluid depend on a number of parameters including the properties of the base liquid and the dispersed phases, particle concentration, particle size and morphology, as well as the presence of dispersants and/or surfactants. This highlights the importance of nanofluids formulation. Two methods can be used for the formulation, namely top-down through size reduction and bottoms-up through simultaneous production and dispersion of nanoparticles. Examples of the bottom-up method include condensation of metal vapor (Eastman et al., 2001) and precipitation of nanoparticles in liquid (Zhu et al., 2004). Eastman et al. (2001) formulated nanofluids by exposing metal vapor on the surface of a moving low vapor pressure liquid. Interactions between the metal vapor/metal particles and the liquid resulted in the formation of nanofluids. The chemical formulation method by Zhu et al. (2004) utilised a chemical reaction inside a microwave oven to produce suspensions of copper nanoparticles.

Compared with the bottoms-up approach, dispersing dry particles into liquids has been more frequently used by researchers working on nanofluids. Various types of nanoparticles with sizes considerably smaller than 100 nm are now commercially available in dry form. However, almost all of these nanoparticles are in the form of agglomerates a dimension much larger than primary particle size due to the nature of the manufacturing process, handling and/or storage. In the dispersed state the actual particle size and its distribution are generally quite different from that of the particles before dispersion. It is essential to disperse these large agglomerates to their primary particle size to make a stable dispersion. The process, by which particles are dispersed in the liquid, plays a critical role for the dispersion under thermal and hydraulic conditions imposed on it. The equipment used in dispersing dried nanoparticles includes ultrasonic bath (Xuan and Li, 2000; Das et al., 2003), magnetic stirrer (Xie et al., 2002), high-shear mixer (Pak and Cho, 1999), homogenizer and bead mill (Way, 2001). The processing time and intensity could also significantly influence the dispersion effect. The weak-bonded agglomerates could be broken to the primary particle size by high shearing; however nanoparticles have a high tendency to re-agglomerate in dispersions due to the nature of attractive London van der Waals force among particles. So electrostatic repulsion force and/or steric hindrance are required to overcome the attractive forces and form stable dispersions. Some surfactants and/or dispersants were often used during the formulation of nanofluids to enhance steric barriers between nanoparticles, therefore to stabilize the nanofluids. However cares must be taken as these surfactants/dispersants could have significant influence on the flow and heat transfer, and some also may fail under high temperatures (Wen and Ding, 2004a). Electrostatic method is therefore preferred for nanofluids applications in some situations.

In this work, TiO_2 nanofluids were formulated through the dispersive method. Distilled water was used as the host liquid and TiO_2 nanoparticles came from a commercial company (Nanophase Technologies Corporation, USA). It is made from chemical vapor deposition method and has a particle density of 4200 kg/m^3. Fig. 1 shows an SEM image of the TiO_2 nanoparticles. It can be seen that the primary nanoparticles are spherical in shape and approximately 30–40 nm but in the form of agglomerates. These agglomerates have to be broken in order to produce stable nanofluids. For doing so, a high-shear homogenizer (Ultra-Turrax T25, IKA) was used to process suspensions. The homogenizer had a gap of 0.5 mm between the rotor and the stator and the rotor rotated at a speed up to 24,000 rpm thus providing a shear rate up to 40,000 s^{-1}. The stabilization of nanoparticles in water was realised by using the electrostatic method. A considerable amount of preliminary effort was made to investigate the stability of TiO_2 nanofluids with different solids concentrations under various pH conditions. Nitric acid (HNO_3) and sodium hydroxide (NaOH) were used to adjust the pH value. A Malvern Nano ZS device with an auto-titrator (Malvern Instruments, UK) was used to measure the Zeta potential and its dependence on the pH. Fig. 2 shows the Zeta potential as a function of pH for 0.024 vol% TiO_2 nanofluid. It can be seen that the iso-electrical point (IEP) of the TiO_2 particles is approximately 6.5. This agrees well with published IEP data of non-surface modified Rutile TiO_2 particles. Theoretically, the further away from the IEP, the more stable the suspension given other conditions. As a consequence, nanofluids were formulated at pH = 3 in this work.

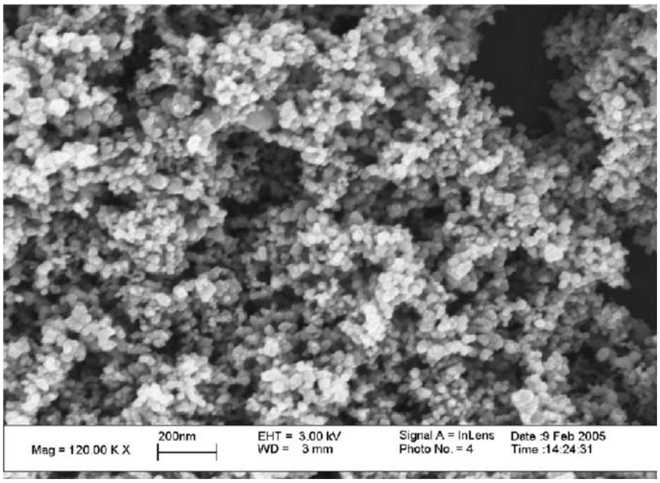

Fig. 1. SEM picture of TiO_2 nanoparticles.

The typical nanofluids formulation process involved: (1) Thorough cleaning of the bottles in ultrasonic bath using

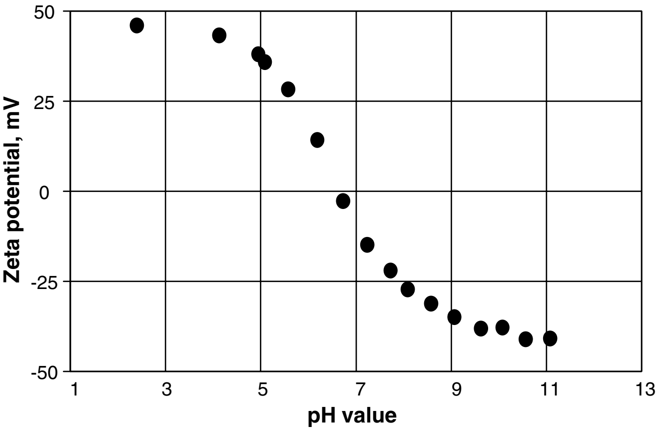

Fig. 2. Zeta potential of TiO_2 nanoparticles (0.024 vol% TiO_2 in distilled water).

an ultrasonic cleaner. (2) Adjusting the pH of the base liquid to pH = 3. (3) Dispersing the required amount of nanoparticles into the bottle of base liquid while keeping the ultrasonic bath switched on. (4) Ultrosonification of the bottles containing dispersion for ~15 min. (5) Shear-mixing of the dispersion under the homogenizer for 30–180 min. The effectiveness of the method for nanofluids preparation was characterized by particle size distribution using a Malvern Nanosizer. Fig. 3 shows the average particle size as a function of high shear processing time. It can be seen the average size decreased rapidly with processing time in the first 50 min after which the dependence levels off. The processing time for all nanofluids preparation was therefore set at 50 min. As shown in Fig. 3, the average particle size is approximately 170 nm. This is much larger than the nominal size of 34 nm provided by the manufacturer and the estimated size from the SEM image (Fig. 1). There are a few reasons for the larger measured average particle size. One is the existence of small agglomerates as mentioned above, especially those particles being closely coalesced or sintered together during manufacturing process. The other is associated with the measurement method. The Malvern Nanosizer measures the hydrodynamic size according to the Stokes–Einstein equation, which is expected to be larger than the actual size. Another possible reason may be due to the low pH value used in the formulation of nanofluids, which produces a thick electric double layer. Further work on this is needed but beyond the scope of this paper. It is to be noted that the above characterization work is necessary, as this will provide information of samples we works on, for which, as mentioned above, little is provided in most published studies on nanofluids.

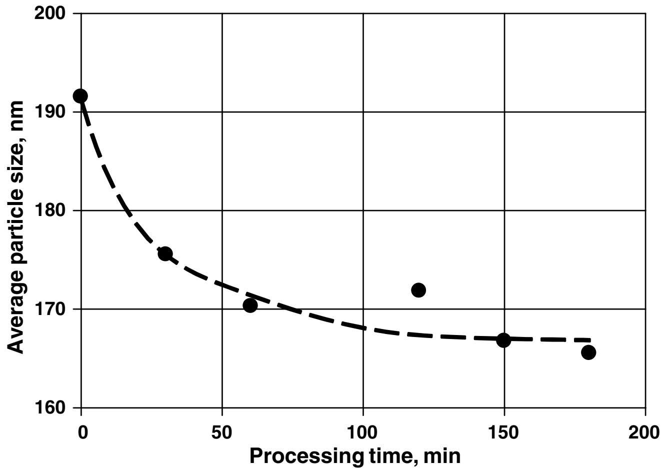

Fig. 3. Average particle size as a function of high shear processing time (0.012 vol% TiO_2 in water, pH = 3).

Despite the use of the measures described above, a very small amount of sediments was still found at the bottom of the container after a few hours, which were believed to be agglomerates that could not be broken down by the methods used in this work. To overcome this, the uniform suspension on the top part of the vessel was carefully transferred to another vessel for natural convective heat transfer experiments. The dispersion was found to be very stable and could last for at least a couple of weeks without visually observable sedimentation.

2.2. Natural convective heat transfer experiment

The experimental system is shown schematically in Fig. 4. It consisted of two horizontally positioned aluminum discs of diameter 240 mm and thickness 10 mm (C and D) separated by a 10 mm gap through a short insulated PTFE cylinder (F). A silicon rubber flexible heater (H) purchased from Watlow UK was closely attached on the lower surface of the bottom disc. The heating element was covered by a thick insulation material (E) to obtain a constant heat flux condition. Electricity supply to the heating element was through a DC power supply (J). The DC power supply was of type TTi Ex752 m (RS, UK), which was adjustable and had a maximum power of 300 W. The upper surface of the top aluminum disc was exposed to the open space and cooled by compressed air. The gap between the aluminum discs was filled with nanofluids for convective heat transfer experiments. The filling was through the filling vessel (A), which was raised approximately 100 mm above the lower surface of the top aluminum disc (C). The filling process was monitored by an

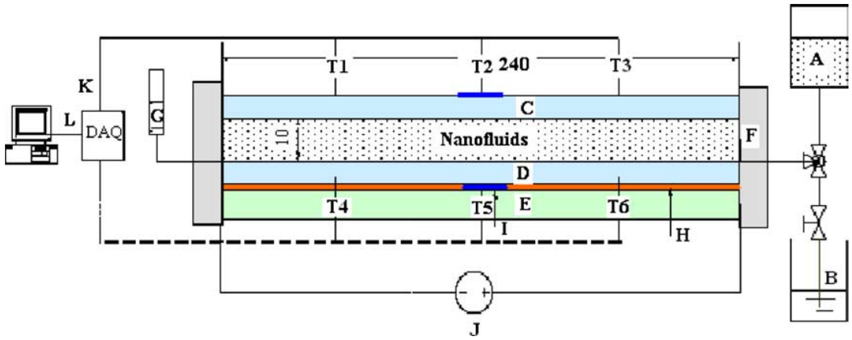

Fig. 4. Experimental setup: (A) filling vessel, (B) discharging vessel, (C) top aluminium plate, (D) bottom aluminium plate, (E) insulation material, (F) insulation cover, (G) level indicator, (H) heating element, (I) heat flux sensor, (J) DC power supply, (K) data acquisition system (DAQ), (L) computer.

indicator G, connected to the test region. A discharge vessel (B) was used to collect nanofluids and rinsing liquid (distilled water). The filling and the discharging vessels and the test region were connected through a three-way valve. Six type J thermocouples (T_1–T_6) and two surface heat flux sensors (I) were mounted on the aluminum surfaces to measure temperatures and heat fluxes. The thermocouples and heat flux sensors were connected to a data acquisition system (K), which was then interfaced to a PC (L) through a NI PCI-6052E board. A SCXI-1102 32-channel thermocouple amplifier (National Instrument, UK) was used to achieve high accuracy temperature measurement during heat transfer experiments. A Labview software was used for system configuration and control.

A typical experiment involved cleaning of the test region by distilled water many times followed by filling with the nanofluids. Great care was needed during the filling process to prevent the formation of gas bubbles in the vessel. Compressed air was switched on and adjusted to cool the upper surface of the upper disc. Heating and data requisition were then started until steady state was reached. Both transient and steady state signals from thermocouple and heat flux sensors were collected. The heat flux was obtained from the input voltage and resistance of the heater by

$$q = U^2/(RA) \tag{1}$$

where U is the voltage, R is the resistance of the heater, and A is the surface area of the heater, which is the same as the surface area of the bottom aluminum disc (and also that of the top disc). At the steady state, the heat diffusion equation was adopted to obtain the surface temperatures of both the upper surface of the bottom disc (T_{ub}) and the lower surface of the top disc (T_{lt})

$$T_{\mathrm{ub}} = T_{\mathrm{mh}} - qd/k_w, \quad T_{\mathrm{lt}} = T_{\mathrm{mc}} + qd/k_w \tag{2}$$

where T_{lb} is the temperature of the lower surface the bottom disc, T_{ut} the temperature of the upper surface of the top disc, d is the thickness of the discs, and k_w is the thermal conductivity of the discs. The heat transfer coefficient, h, was calculated by

$$h = q/(T_{\mathrm{ub}} - T_{\mathrm{lt}}) \tag{3}$$

All the thermocouples were calibrated prior to experiments, which gave an accuracy of 0.2 K. Heat loss of the experimental setup was measured by running steady heat conduction experiments with top heating and bottom cooling, which was smaller than 7% under the conditions of this work. All the temperature data were recorded throughout the experiments with a DAQ rate of 200 fps but only some intermittent points were used for figures shown below.

3. Results and discussion

3.1. Experiments with distilled water

Initial experiments were conducted with distilled water to check the performance of the system. Fig. 5 shows the results presented in the form of Nusselt number (Nu) as a function of the product of the Grashof number (Gr) and Prandtle number (Pr), defined respectively as

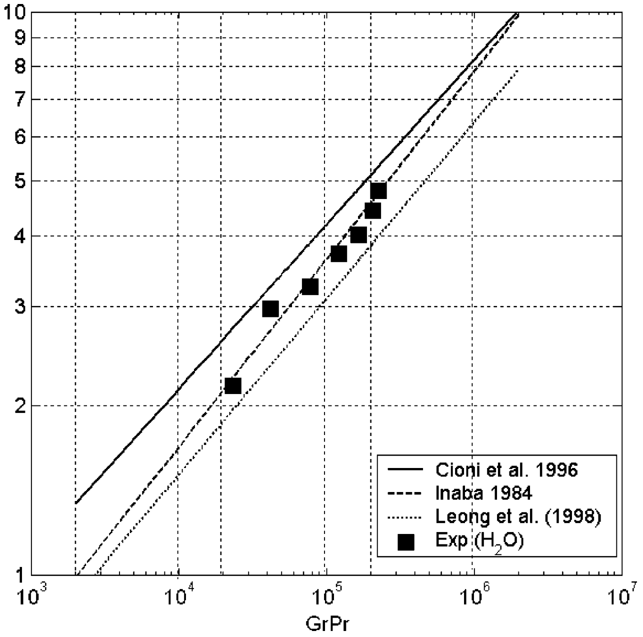

Fig. 5. Comparison between experiments and published results for natural convection of water.

$$Nu = hd_{\mathrm{g}}/k_{\mathrm{f}}, \quad Pr = v_{\mathrm{f}}/\alpha, \quad Gr = g\beta\Delta T d_{\mathrm{g}}^3/v_{\mathrm{f}}^2 \tag{4}$$

where d_{g} is the separation between the two discs, k_{f} the thermal conductivity of the fluid, v_{f} the kinematic viscosity of the fluid, α the thermal diffusivity of the fluid, g the gravitational acceleration, β the volume expansion coefficient of the fluid and ΔT the temperature difference between the two disc surfaces. The product of Gr and Pr numbers is also termed as the Rayleigh number defined as $Ra = g\beta\Delta T d_{\mathrm{g}}^3/(v_{\mathrm{f}}\alpha)$. There are many empirical correlations in the literature for natural convection heat transfer with most of them taking the following form:

$$Nu = cRa^n \tag{5}$$

where c and n are the empirical constants. Included in Fig. 5 are predictions of the correlations by Inaba (1984), Leong et al. (1998) and Cioni et al. (1996). It can be seen that the experimental results lie within the predictions by the three correlations but agree best with the Inaba correlation over the range of Rayleigh number used in this work. The maximum discrepancy is ~17% for a Rayleigh number of 40,000, which was thought to be due to the side-wall effect and different aspect ratios.

3.2. Transient temperature and heat transfer coefficient

Examples of transient temperature signals for different concentration of TiO_2 nanofluids are shown in Fig. 6 under a heat flux of $q = 190\ \mathrm{W/m^2}$. To make a quantitative comparison, all the experiments were performed under the same

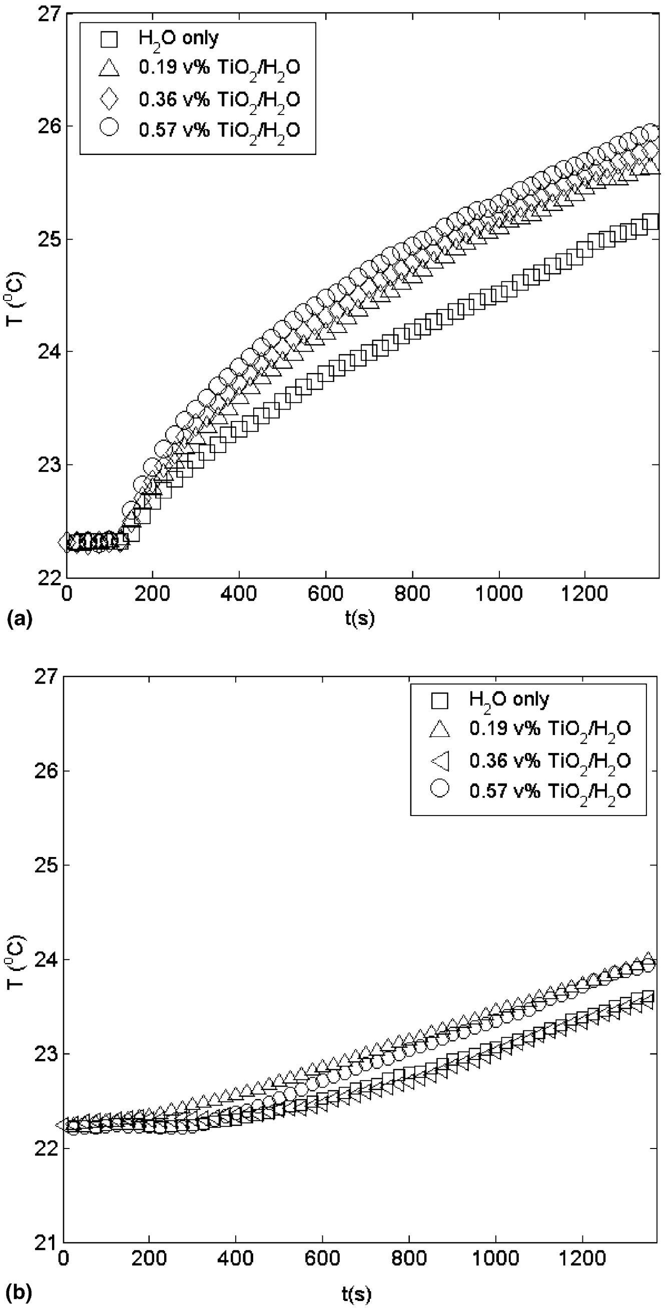

Fig. 6. Surface temperature histories: (a) heating surface (T_h); (b) cooling surface (T_c).

conditions and temperature signals were acquired every 5 s. It is noticeable that the temperature increases quite smoothly and without any initial temperature oscillations as observed by Okada and Suzuki (1997) and Kang et al. (2001) for slurries comprising of micrometer-sized particles. The time to reach steady state was also much shorter. This indicated that the sedimentations responsible for the initial instabilities in micro-sized particles slurries were not a problem for nanofluids. Similar smooth temperature signals were also observed by Putra et al. (2003) for CuO and Al_2O_3 nanofluids in a horizontal vessel. However contrary to previous belief that nanofluids should enhance heat transfer through increased effective thermal conductivity, the heating surface temperature was found to increase with nanoparticles concentrations. The bottom temperatures were found to increase rapidly following the switch-on of the heating power and approached to a nearly linear increase after about 600 s. Little difference, however, was found for the transient top surface temperature as shown in Fig. 6(b). The temperature difference between the heating and cooling plates was therefore increased with nanoparticle concentrations, Fig. 7. A nearly constant temperature difference was reached after ~1200 s. With only a 1.5 K difference found for the pure liquid, the temperature difference increased with the concentration and reached 2.3 K for a 0.57 vol% TiO_2 nanoparticle concentration. Similar phenomena were also observed for all the other cases, which clearly showed that the addition of nanoparticles systematically decreased heat transfer coefficients.

The variations of Nusselt and Rayleigh numbers as a function time are illustrated in Fig. 8. The base liquid properties, i.e., thermal conductivity, heat capacity and viscosity, were used in deriving the dimensionless numbers, whose influences are discussed in next section. It can be seen that the Nusselt number decreases continuously with time while the Rayleigh number increases during the heating period. As a result, the Nusselt number decreases with the Rayleigh number during the transient heating period, Fig. 9. Similar phenomena were also observed by Tso et al. (2004) and Bhowmik and Tou (2005) for cooling of electronic chips through natural convection in a vertical rectangular channel.

The steady state natural convective heat transfer data for the distilled water and three concentrations of nanofluids are depicted in Fig. 10. The correlation of Inaba

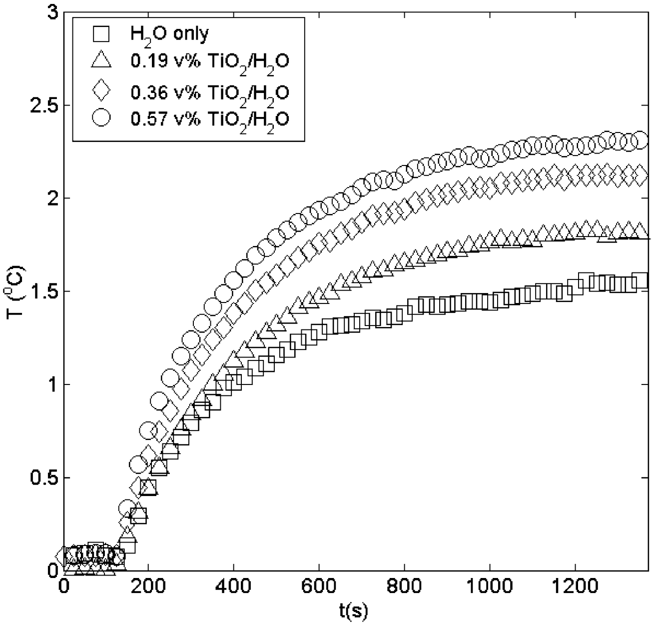

Fig. 7. Temperature difference between the heating and cooling surfaces ($T_h - T_c$).

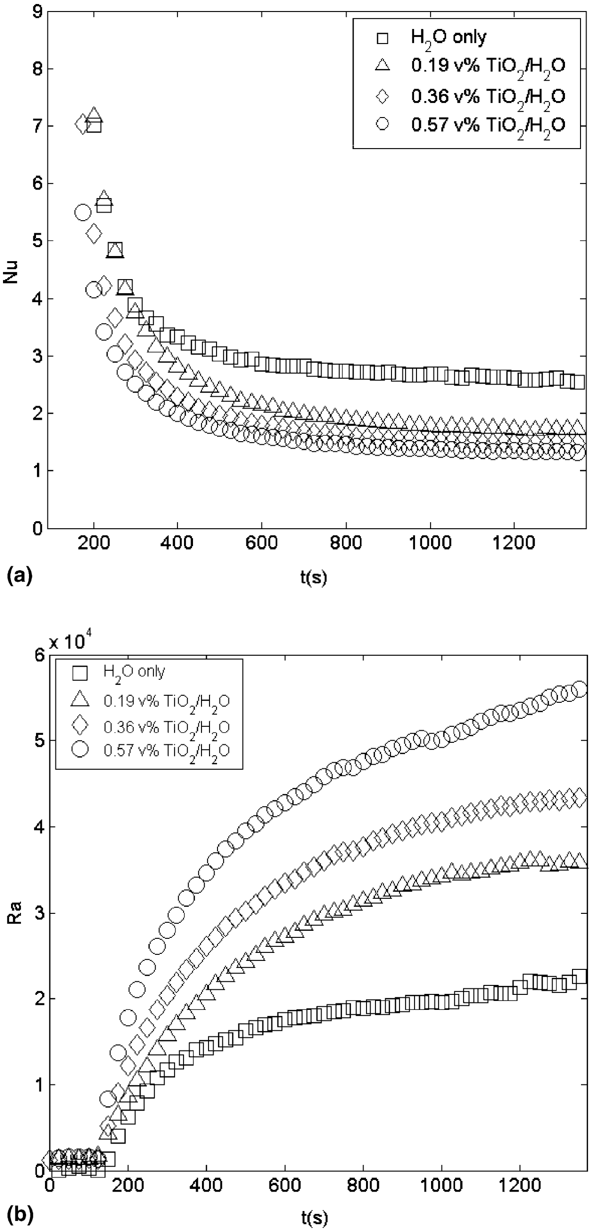

Fig. 8. Nusselt and Rayleigh numbers as a function of time (heat flux of 190 W/m^2). (a) Nusselt number and (b) Rayleigh number.

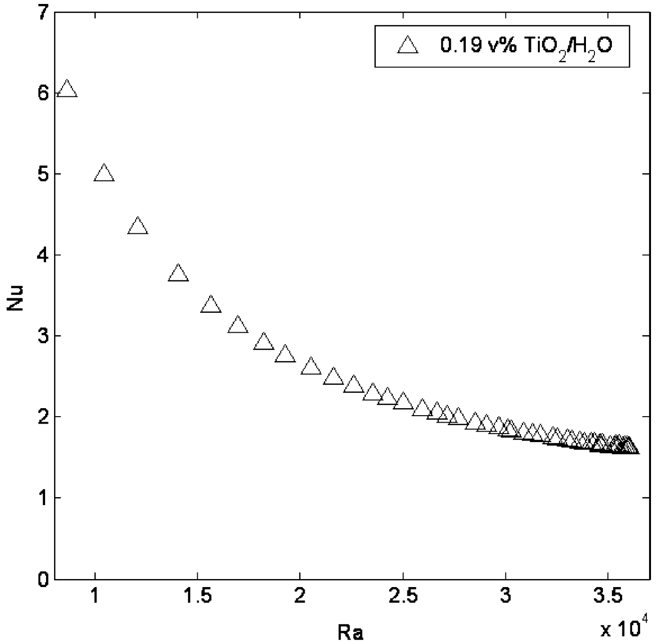

Fig. 9. Transient Nusselt vs Rayleigh number ($q = 190$ W/m^2).

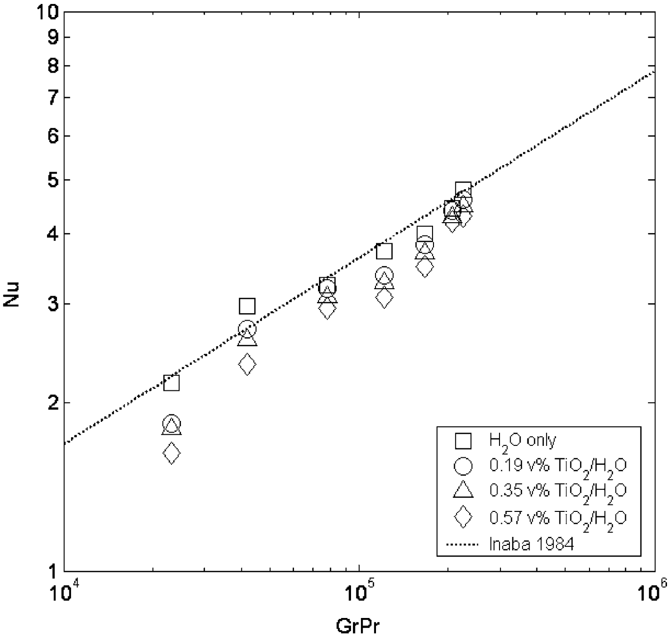

Fig. 10. Steady state heat transfer results.

(1984) is also given in the figure for comparison. Apparently, the Nusselt number decreases with nanoparticle concentration, particularly at low Rayleigh numbers. Possible reasons for this unexpected result will be discussed in the following section.

3.3. Discussions

The observation that the presence of nanoparticles deteriorates the natural convective heat transfer is somewhat paradoxical. The results are different from that numerical simulations by Khanafer et al. (2003) for natural convective heat transfer behaviour of nanofluids in a two-dimensional horizontal enclosure. However the experimental results do agree with the observations by Putra et al. (2003) who also observed decreased natural convective heat transfer coefficient for aqueous CuO and Al_2O_3 nanofluids inside a horizontal cylinder. The simulations of Khanafer et al. (2003) were based on some ideal assumptions of (a) nanofluid was Newtonian, incompressible and the flow was in the laminar regime; (b) nanoparticles were uniform in shape and size; (c) there was no slip between liquid and particle phases in terms of both velocity and temperature; and (d) nanofluids had constant thermophysical properties except for density variation that gave rise to the buoyancy.

The assumption of (c) and (d) are very difficult to be satisfied for real nanofluids. Although nanofluids behave more like pure fluids than suspensions of large particles, fundamentally speaking, they still exhibit two-phase characteristics that need to be considered for thorough understanding of nanofluids heat transfer.

A number of possible mechanisms have been proposed for the enhancement of effective thermal conductivity (Keblinski et al., 2002), which included Brownian motion effect, interfacial liquid layering, ballistic transportation of energy carriers within nanoparticles and formation of nanoparticle structures through fractal, clustering and networking. For nanofluids application under forced convective conditions, possible contributions to the complex behaviour of the nanofluids may include the Brownian motion, particle rotation and its associated micro-convection, particle migration and the resulting non-uniform property profile, and suppression or interruption of the boundary layer (Wen and Ding, 2005). The mechanisms associated with nanofluids under natural convective heat transfer conditions are expected to be quite different. While Putra et al. (2003) ascribed the possible reasons of deterioration to the effects of particle–fluid slip and sedimentation of nanoparticles, which might still be the possible reasons for the present work; other reasons may come into play as analyzed below.

3.3.1. Property influence

From a macroscopic view, the properties of a homogenous nanofluids that affect the heat transfer behaviour include heat capacity, thermal conductivity, density and viscosity. For dilute dispersions as used in this work (less than 1% by volume), the heat capacity of the mixtures does not differ too much to the base liquid. However there could be some big difference in terms of the effect of effective thermal conductivity and viscosity. A typical set of thermal conductivity results is shown in Fig. 11 for measurements at 22 °C, where k is the thermal conductivity of nanofluids. It can be seen that the effective thermal conductivity of nanofluids increases approximately linear with particle volume fraction under the conditions of this work, and an enhancement of ~6.4% is achieved at a particle concentration of ~0.8 vol%. Together shown in Fig. 11 are predictions by the MG (Choy, 1999), the Bruggeman (Choy, 1999), the DEMT (Garboczi and Berryman, 2000) and the HC (Hamilton and Crosser, 1962) models developed for macroscopic systems, which obviously fail to predict the measured results. This observation is consistent with many studies (i.e., Choi, 1995; Eastman et al., 1996; Xuan and Li, 2000; Xie et al., 2002; Wang et al., 2003; Wen and Ding, 2004a,b). However the heat transfer performance of the nanofluids was not solely decided by the effective thermal conductivity. Other parameters, such as particle size and concentration distribution, and the apparent viscosity, may also become important. For example, Tseng and Lin (2003) investigated the rheological behaviour of aqueous TiO_2 nanofluids, and proposed a simple correlation for the relative viscosity, η as below,

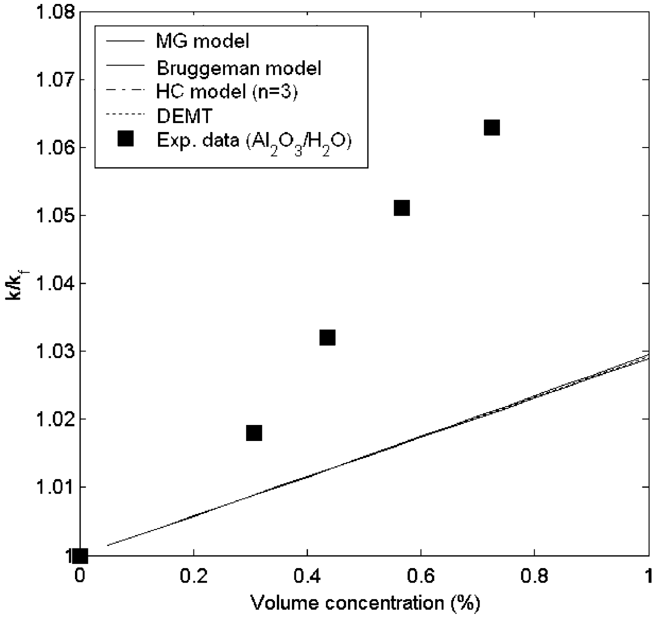

Fig. 11. Measured effective thermal conductivity and comparison with some macroscopic models.

$$\eta = \mu_e/\mu_l = 13.47e^{35.98\phi} \qquad (6)$$

where μ_e and μ_l are the effective and base liquid viscosities respectively, and ϕ is the volume fraction. The big exponents imply significant viscosity increase for even a small increase of particle concentration. The addition of nanoparticles may also influence the surface wettability and other surface parameters, whose effect is still not clear.

The increase in the effective thermal conductivity and viscosity of nanofluids reduces the Nusselt number and Rayleigh number values under similar conditions. The relative importance, as shown in Fig. 8, is difficult to estimate. There may exist an optimum concentration that gives the best performance. However no matter what are these values, it could not change the observation that the addition of nanoparticles decreases heat transfer coefficients as shown in Fig. 7, as the temperature difference increased with nanoparticle concentrations under the same heat flux conditions.

3.3.2. Convection by concentration difference

Besides the convection induced by liquid due to the density variation, which is often approximated by including the Boussinesq equation into the Navier–Stokes equation, the big density difference between solid nanoparticles and liquid may also induce convection. Such convection may become significant especially as nanoparticles aggregated in one place and produced depletion zones. Macroscopical convection could therefore be induced, which may be coupled with the Boussinesq convection and makes the flow and heat transfer behaviour of nanofluids more complex. Due to the strong non-linear coupling, direct analytical solution of such phenomena is always difficult; full numerical simulations are needed to further reveal the details.

Although Khanafer et al. (2003) simulated natural convective heat transfer of nanofluids, the model considered homogeneous distribution of nanoparticles, and without considering either detailed thermal conductivity or the rheological behaviour of nanofluids. Their predictions do not therefore apply to the case of this work.

3.3.3. pH influence

Electrostatic stabilization method was used in formulating the stable TiO_2/H_2O nanofluids used in the experiments. As a consequence, the pH value of this nanofluids was adjusted to be very low, $pH = 3$. While there are still some arguments whether pH values could affect the effective thermal conductivity of the nanofluid, the low pH value does create strong surface charges on the nanoparticles to prevent aggregations and keep the nanofluids stable. From a microscopic point of view, it is the particles motion and interactions with surrounding liquid and other particles that enhance the heat transfer when compared with the single phase liquid conduction only. Such interparticle interactions arising from the increased electrostatic charges may be responsible for the deteriorated heat transfer observed. It is therefore also possible that some degree of interparticle interaction is required to observe enhanced heat transfer. However more experiments and analytical works need to clarify these issues, which will be a next target.

3.3.4. Particle–surface interactions

Natural convective heat transfer is affected by a number of processes in parallel and/or series, including unsteady-state heat conduction through the heating wall, conduction within the boundary layer and its development, as well as convection due to the variation of liquid density and the density difference between the nanoparticles and the liquid. As a possible process, nanoparticles, if not specially treated, will form a layer on the surface, which might significantly change surface properties. Such phenomenon is more prominent for unstable nanofluids, where agglomerates may settle down and deposit on the surface. The agglomerates may stick/sinter on the heating surface and generate a fouling effect. Although the melting point of the bulk material is high, that of nano-sized particles may be considerably lower hence the possibility of sintering. The layer of nanoparticle agglomerates deposited on the heating surface may generate an extra thermal resistance to the surface and prevents the direct contact of liquid with the surface and possibly decreases the heat transfer.

4. Conclusions

This paper is concerned with the formulation of aqueous based nanofluids and its application under natural convective heat transfer conditions. It shows that very stable titanium dioxide/water nanofluids could be formulated through the mechanical shear mixing and electrostatic stabilization. Both transient and steady heat transfer coefficients were obtained for different concentrations of nanofluids under natural convective conditions. The nanofluids are found to decrease the natural convective heat transfer coefficient; such deterioration increases with nanoparticle concentrations. Possible reasons/mechanisms attributed to such behaviour are discussed, including the convection induced by concentration difference, particle–surface and particle–particle interactions, and modifications of the dispersion properties. Further experimental and theoretical works are being carried on to identify the exact causes.

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