Convective Heat Transport in Nanofluids

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1 Introduction

We have assembled an interdisciplinary team to investigate heat transfer by convection in nanofluids. The concept of a nanofluid has been advanced by S. Choi 1995[1] who showed substantial augmentation of heat transported in suspensions of copper or aluminum nanoparticles in water and other liquids. Nanofluids are a new kind of fluid; they are dispersions of nanoparticles in liquids that are permanently suspended by Brownian motion. By using different solvents and particles we hope to create composite fluids of widely variable and perhaps completely new properties. Here we draw attention to the difference between nanofluids and nanoparticles; they are different materials with different applications. Though our general goal is the creation and study of transport properties of nanofluids this proposal focuses on heat transport. The heat transport properties of nanofluids depend on the thermal properties, concentration size and shape of suspended nanoparticles. At Minnesota we propose to combine our great strengths in heat transfer, multiphase fluid mechanics and particle technology in a focused study of convective heat transport in nanofluids.

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[1] Choi, U.S., 1995, *Enhancing Thermal Conductivity of Fluids with Nanoparticles, Developments and Applications of Non-Newtonian Flows*, D.A. Siginer and H.P. Wang, eds, FED-Vol. 231/MD-Vol. 66, ASME, New York, pp 99-105.

2 Nanofluids

A nanofluid is a more or less uniform dispersion of solid particles with small diameters measured in nanometers. These particles are suspended by Brownian motions and when they are in equilibrium with no flow they are distributed in a balance between buoyant weight and thermal agitation (Brownian motions). In fact this kind of equilibrium does not generate a uniform dispersion; instead the concentration of small particles of radius a will decrease exponentially with height according to the well-known theory of Albert Einstein. This variation can be nearly uniform for very small particles in devices of laboratory dimension. The balance between the small fluctuations and buoyant weight can be expressed by the gravitational height

$$l = \frac{kT}{(\rho_p - \rho_l) g \frac{4}{3}\pi a^3}$$

where k is Boltzmann's constant, T is in degrees Kelvin, ρ_p is particle density and ρ_l is fluid density. This quantity appears in the exponential distribution of equilibrium concentration

$$c(z) = \bar{c} \exp\left(-\frac{m}{l} z \right)$$

where m is the compressibility of the suspension, a quantity related to the osmotic pressure that swells the bed (see Phan *et al* 1996[2]). This expression shows that the concentration is nearly uniform when l is large. For nanoparticle, l is in 10's of meters even for copper densities $\rho_p \approx 8$ gm/cms. Larger particles with a's in the submicron but not nanometer range may be very nearly uniformly suspended by Brownian motions.

Piazza, Bellini and Degiorgio 1993[3] point to discrepancies between measured and theoretical values of the gravitational length with considerably larger values in the experiments.

As far as we know, no direct experiments have been carried out for the critical size of particles below which particles can be permanently suspended by Brownian motion to within tolerance.

3 Critical size

A good candidate for a dimensionless length is the diameter of a small particle below which the special effects of nanoparticles become evident. In the most primitive description of two-phase flow models of solid liquid systems the effects of the particles is accounted for in terms of the solids fraction ϕ. The size and shape of particles is ignored. Let us focus on size. Suppose we have predicted some behavior of the solid-liquid system in terms of the solids fraction ϕ; then we fix ϕ and reduce the size of the particles at a fixed ϕ. Naturally some size effects are expected, but they may be and frequently are small. If nanoparticles have a big effect we will begin to see large departures at a fixed ϕ in the behavior of our solid-liquid system when the size d of the particles is reduced past some perhaps vaguely defined value d_n. It is this value d_n that we would

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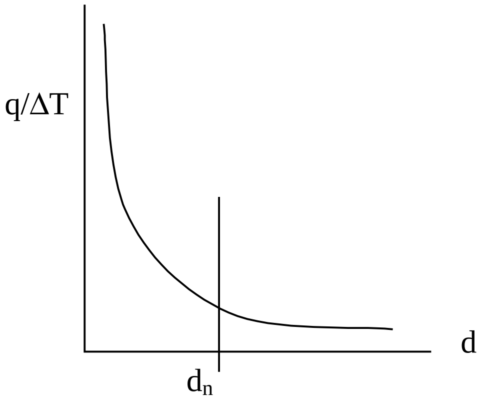
[2] S-E Phan, WB Russel, Z Cheng, J Zhu, PM Chaikin, JH Dunsmuir and RH Ottewill 1996. Phase transition, equation of state, and limiting shear viscosities of hard sphere dispersions, *Physical Review E*, **54** (6), 6633-.

[3] R Piazza, T Bellini and V Degiorgio, 1993. Equilibrium sedimentation profiles of screened charged colloids: a test of the hard-sphere equation of state, *Physical Review Letters*, **71**(25), 4267-.

use for our dimensionless length. In section 4 we argue that for nanoparticles convection the critical size d_n is the largest size at which heavy particles go into a colloidal dispersion. The critical value d_n could appear, say as the size of particles at which sharp departures of the heat q transported across a fluid layer by a temperature difference ΔT first appears; as in the diagram below.



The effects of size variations at a fixed ϕ may be smooth or sharp. We would expect that when new phenomena like short range forces or self-assembly are introduced at the nanoscale, the variation of the behavior at a fixed ϕ will be very sharp; in the case where the main effects are merely due to an increase of area at a fixed volume the variation will be smooth. The two–phase flow description for this second smooth case may perhaps not be overly complicated.

4 Techniques for preparing nanofluids

The evolution of reliable techniques for creating stable and well-dispersed suspensions of nanoparticles is crucial to the success of our research. To test new ideas for cool applications of nanofluids we need to possess reliable techniques for making them. To create a nanofluid the particles should be small enough to suspend by Brownian motion and the particles must be protected against aggregation by electric charges, protective coatings or by other means.

Brownian motions are intrinsically dispersive and in the absence of aggregative effects would produce diffusion of nanoparticles along lines suggested by the miscible liquids analogy. In fact aggregation of particles does occur, depending on the particles and the solvent environment; these problems of aggregation are particularly severe at volume fractions over 20% by volume or less. New techniques for suppressing aggregation are greatly desired.

There is a small literature on methods of preparation of nanofluids pioneered by the group of S. Choi. They use two techniques to make nanofluids, described as "the single-step direct evaporation method, which simultaneously makes and disperses the nanoparticles directly into the base fluid, and the two-step method, which first makes nanoparticles and then disperses them into the base fluid. Although the two-step techniques works well for oxide nanoparticles, it is not as effective for metal nanoparticles. For nanofluids containing high conductivity metals, it is clear that the single-step direct evaporation technique is preferable to the gas-condensation processing."

Electrohydrodynamic Spraying System (ESS). The electrohydrodyamic Spraying System (ESS), or electrospray, can be used to produce nanoparticles from a solution of desirable solute materials or colloidal suspension. Airborne nanoparticles in the size range of 2-100 nm can be generated with a production rate of up to 10 billion particles/s. Figure 2 shows a schematic diagram of the electrospray (Chen and Pui, 1995, 1997[4]). A syringe pump (or pressurized chamber) is used to deliver a solution or suspension to a capillary tube. The liquid exits through the capillary tube will be exposed to a high electric field established between the tip of the capillary (electrically grounded) and the orifice plate (high voltage applied) downstream. The nonuniform electric field causes the liquid meniscus to assume a conical shape (see the photograph in figure 1) established by the balance between the surface tension force and electrical force on the cone. The liquid will be dispensed through the cone-tip in the form of liquid jet of nanometer diameter. The instability of the liquid jet will cause it to be broken up into nanoparticles each carrying a very high charge level on the order of 80-90% of the Rayleigh limit. The highly charged particles of the same polarity (unipolar charge) will cause them to expel and disperse from each others. The "space charge" effect thus prevents aggregation of the nanoparticles. The capillary is typically operated with a CO_2 sheath to reduce the potential for corona discharge at the capillary tip and with a dispersion air to dilute and transport the nanoparticles. These nanoparticles can be directly introduced into the base fluids using Choi's mixing method. The droplet size and the particle generation rate are functions of the liquid feed rate and the electrical conductivity of the spray solution/suspension. We have successfully sprayed 60 different solvents with different solute materials to produce a variety of nanoparticles of desirable materials (Chen and Pui, 1997). Colloidal suspension of metal, oxide and polymer nanoparticles have also been sprayed to form dispersed nanoparticles without aggregation. A dual capillary approach has also been used to provide a sheath liquid of varying conductivity. It will allow the core liquid to be sprayed without having to add foreign substance to alter its conductivity. The sheath liquid can also be made of desirable coating materials for producing coated nanoparticles. We have recently developed a multiple-nozzle approach to increase the throughput of producing nanoparticles by over 1,000 times.

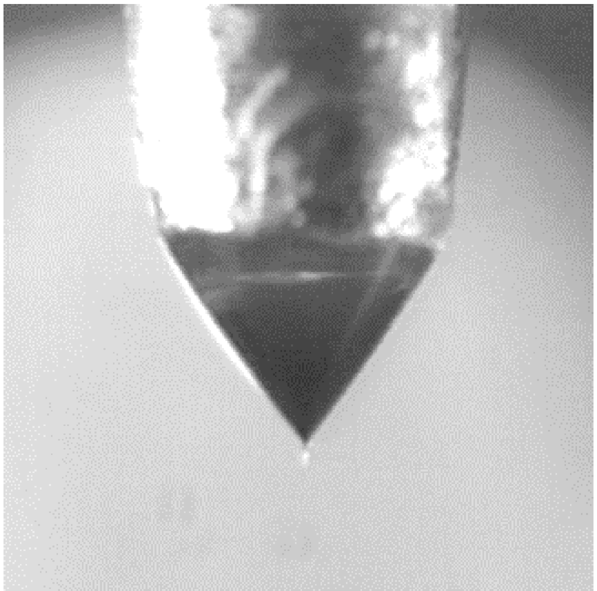


Figure 1. Electrohydrodynamic Spraying System

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[4] Chen, D.R., D.Y.H. Pui, and S.L. Kaufman (1995) J. Aerosol Sci., 26, p963-977.
Chen, D.R. and D.Y.H. Pui (1997)Aerosol Sci. Technol. 27, p367-380.

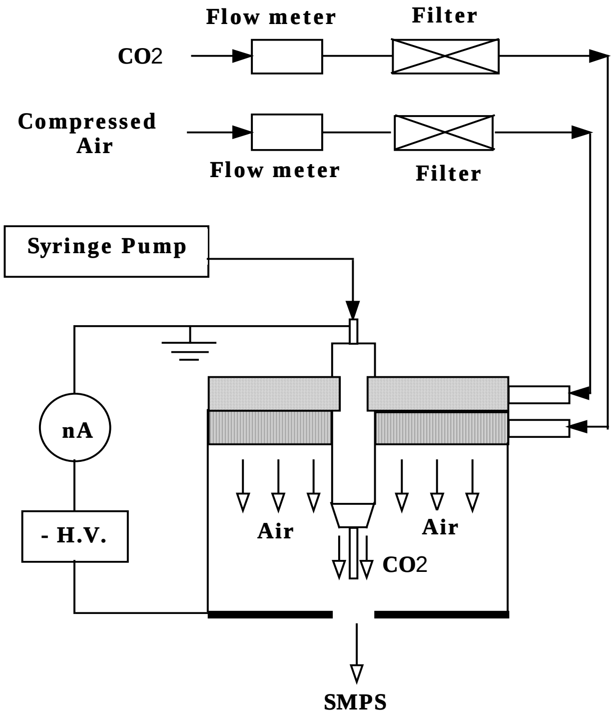

Figure 2. Cone-jet Mode to Produce Monodisperse Nanoparticles

Nanoparticle Differential Mobility Analyzer (Nano-DMA). A Nanoparticle Differential Mobility Analyzer (Nano-DMA) has been developed for measuring and classifying nanoparticles in the size range of 3-100nm. Although the instrument is developed to measure airborne nanoparticles, we have successful electrosprayed highly concentrated colloidal suspensions (20% weight fraction) of Chemical Mechanical Polishing (CMP) powders and measured them for size distribution analysis. Figure 3 shows a schematic diagram of the Nano-DMA and examples of measured size distributions of gold [**~~and sucrose~~] nanoparticles using this instrument (Chen et al., 1998). The particle size is derived from the measured electrical mobility of the nanoparticles. The particle electrical mobility is measured by passing the airborne nanoparticles, "aerosols," in a laminar stream through the concentric cylindrical condenser in which an electric field perpendicular to the direction of aerosol flow is maintained. The perpendicular electric field precipitates the charged particles onto the inner condenser electrode except those particles that enter through a slit near the bottom of the electrode. The Nano-DMA therefore serves as a band-passed filter, only those particles with a known electrical mobility (calculated from the voltage applied to the electrode, the flow rate, and the instrument dimensions) and a narrow mobility range (uniform size particles) will exit the instrument. A particle detector can measure these nanoparticles. By varying the voltage on the condenser and detecting the particle counts, the entire particle size distribution entering the instrument inlet can be measured (see the two sample size distributions in Figure 3 **second sample didn't transfer --Dave). The method is capable of high precision sizing of nanoparticles. We have worked with the National Institute of Standards and Technology (NIST) to certificate 0.1 µm Standard Reference Material 1963 of latex spheres (Kinney et al., 1991) and have recently extended it to smaller nanoparticles. Further, it is shown that the instrument can accurately measure the standard deviation of the narrowly distributed SRM particles (s/Dp = 0.02) (Chen et al., 1999).

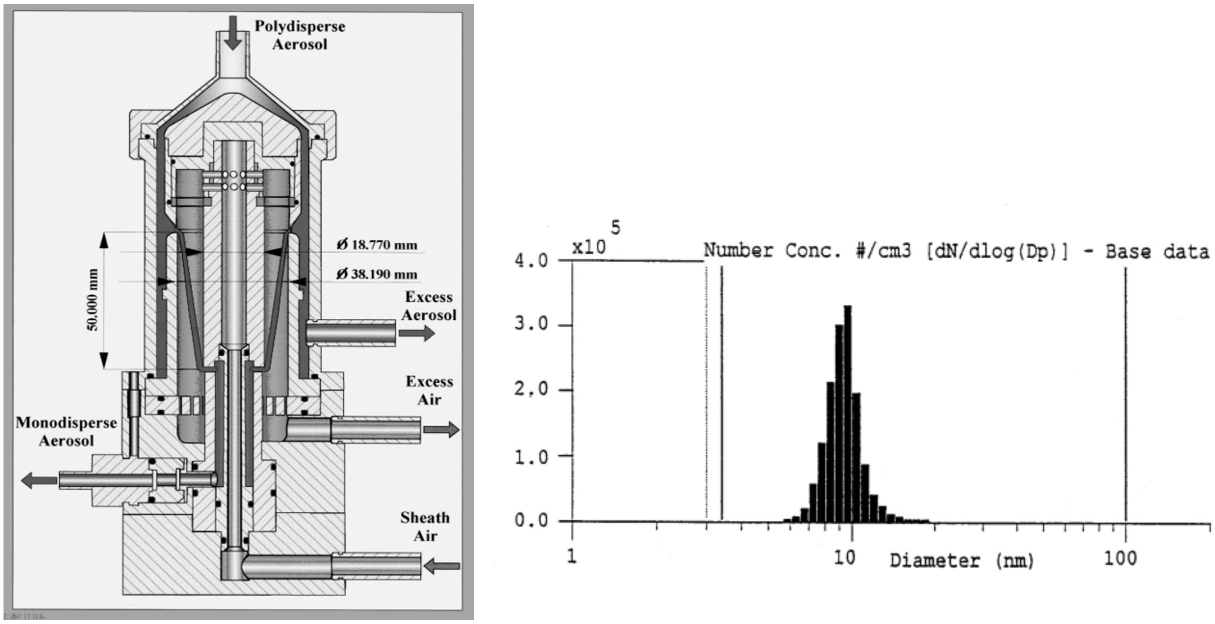

Figure 3. Nano-DMA schematic (left) and 10nm colloidal gold particles.

Chen, D.R. and D.Y.H. Pui (1997) "Numerical Modeling of the Performance of Differential Mobility Analyzers for Nanometer Aerosol Measurements," J. Aerosol Sci., 28, p985-1004.

Chen, D.R., D.Y.H. Pui, D. Hummes, H. Fissan, F.R. Quant and G.J. Sem (1998) "Design and Evaluation of a Nanometer Aerosol Differential Mobility Analyzer (Nano-DMA)," J. Aerosol Sci., 29, p497-509.

Chen, D.R., D.Y.H. Pui, G.W. Mulholland and M. Fernandez (1999) "Design and Testing of an Aerosol/Sheath Inlet for High Resolution Measurements with the DMA," J. Aerosol Sci., 30, p983-999.

Kinney, P., D. Y.H. Pui, G.W. Mulholland and N.P. Bryner (1991) "Use of Electrostatic Classification Method to Size 0.1 µm SRM Particles -- A Feasibility Study," J. Research NIST 96:147-176 (1991).

and here are the titles for electrospray:

Chen, D.R., D.Y.H. Pui, and S.L. Kaufman (1995) "Electrospraying of Conducting Liquids for Monodisperse Aerosol Generation in the 4 nm to 1.8 µm Diameter Range," J. Aerosol Sci., 26, p963-977.

Chen, D.R. and D.Y.H. Pui (1997) "Experimental Investigation of Scaling Laws for Electrospraying: Dielectric Constant Effect," Aerosol Sci. Technol. 27, p367-380.

5 Heat transport and nanoparticles

Various authors, especially S. Choi and his collaborators, have carried out experiments that have demonstrated that suspensions of nanoparticles in fluids results in substantial improvements in heat transport. These papers, which are listed below, interpret the improvements in heat transport in terms of the theory of heat conduction neglecting other modes of heat transport like convection. I think and will argue that the observed increases are due in part to convection in which nanoparticles in colloidal suspension are advected from place to place; these particles are very efficient in transporting heat. In any case we want to study heat transport by convection in fluids with dispersions of nanoparticles.

References that document improved heat transport in fluids with nanoparticles. These all interpret the improvements in terms of heat conduction.

Masuda, H., Ebata, A., Teramae, K., and Hishinuma, N., 1993, Alteration of Thermal Conductivity and Viscosity of Liquid by Dispersing Ultra-fine Particles (Dispersion of g-Al2O3, SiO2, and TiO2 Ultra-fine particles), *Netsu Bussei* (Japan), Vol. 4, No. 4, pp. 227-233.

Choi, U.S., 1995, *Enhancing Thermal Conductivity of Fluids with Nanoparticles, Developments and Applications of Non-Newtonian Flows,* D.A. Siginer and H.P. Wang, eds, FED-Vol. 231/MD-Vol. 66, ASME, New York, pp 99-105.

Artus, R.G.C., Measurements of the Novel Thermal Conduction of a Prophoritic Heat Sink Paste, *IEEE Transactions on Components, Packaging and Manufacturing—Part II*, Vol. 19, No. 3, 1996, pp. 601-604.

Eastman, J.A., Choi, U.S. Li, S., Thompson, L.J., and Lee, S., 1997, Enhanced Thermal Conductivity Through the Development of Nanofluids, Proceedings of the Symposium on Nanophase and Nanocomposite Materials II, Vol. 457, Materials Research Society, Boston pp 3-11.

S. Lee, S. Choi, S. Li and J. Eastman, May 1999, Measuring thermal conductivity of fluids containing oxide manoparticles, *Transactions of the ASME*, **121**, 280-289.

X. Wang, X. Su and S. Choi, June 1999, Thermal conductivity of nanoparticles-fluid mixture. *J. Thermophysics*, **13**(4), 474-480.

S. Choi calls fluids laden with nanoparticles, nanofluids. I would add the condition that the particles must be in colloidal suspension. Choi and his colleagues have carried out experiments on heat transport in systems with CuO nanoparticles in water and Al_2O_3 particles in ethylene glycol and water. They find that the particles enhance the heat transport by as much as 20% and they interpret their result in terms of an improved thermal conductivity k/k_0, see figure 4. A schematic of the transient hot wire experiment used to get the results in figure is shown in figure 2 and cautionary remarks are in the caption.

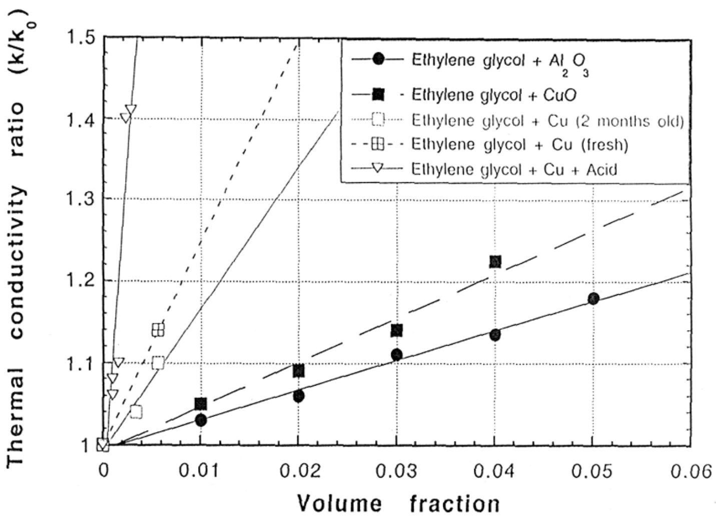

Figure 4. (S. Choi, lecture at U of MN, August 10, 2000) Thermal conductivity measurements using transient hot-wire apparatus.

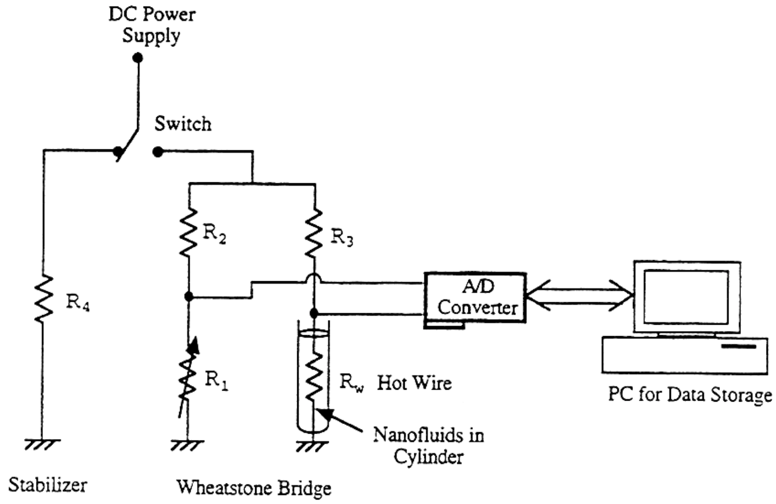

Figure 5. Schematic diagram of transient hot-wire apparatus for measuring thermal conductivities of nanofluids (A/D = analog-to-digital). The formula based on the solution of Fouriers law for conduction from line source into an infinite (not cylindrical) region is

$$T(t) - T_{ref} = \frac{q}{4\pi k} \ln\left(\frac{4K}{a^2 C} t \right) ,$$

where T(t) is the temperature of the wire in the fluid at time t, T_{ref} *is the temperature of the cell, q is the applied electric power, k is the thermal conductivity, K is the thermal diffusivity of the fluid, a is the radius of the wire, and ln C = g, where g is Euler's constant. This formula works very well at very early times in organic liquids before convection sets in. At later times there convection is important and is probably more important in nanofluids. It is impossible to measure thermal conductivity of electrically conducting fluids because current flows through the liquid as well as the wire. In the experiment a platinum wire was coated with an insulator to solve this problem but the solution may not have worked perfectly.*

Wang, Xu and Choi [1999] did a study of heat transport in a nanofluid heated from above as shown in figure 6. The fluid does not fill the cell; there is a free surface. Though fluids heated from above do not introduce gravitational (Bernard) convection they may create convection as an interfacial effect due to nanoparticle concentration differences (Maragnoni convection). We do not know if nanoparticles are amphiphilic but new effects on convection also may occur due to the difference in the coefficient of thermal expansivity of the water and the nanoparticles. It is of interest also to look at the effects of nanoparticles in fluids heated from below.

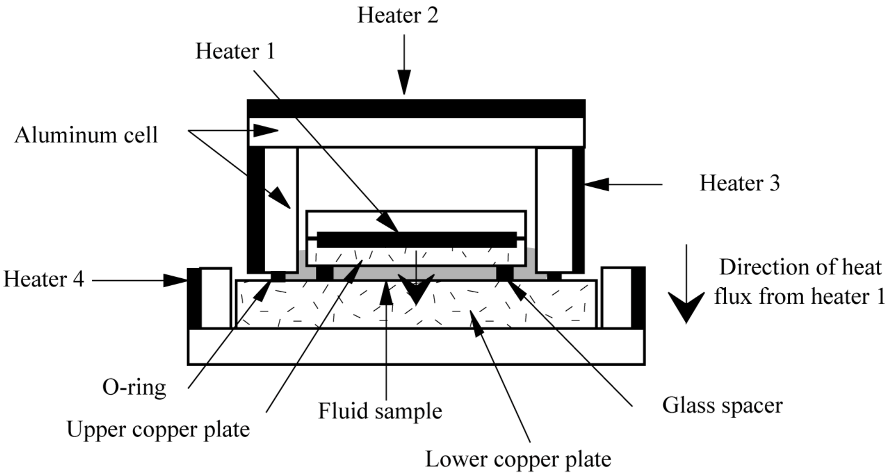

Figure 6. (Wang, Xu, Choi [1999]). The liquid does not fill the space below the copper plate and it can move freely. The data is steady and processed with Fourier's law for conduction.

The focus of heat transport studies in nanofluids is the determination of a size effect at fixed volume fraction. Choi 2000 has demonstrated such an effect in his experiments on heat conductivity; the conductivity increases by 20% as the size of the particle is increased from 38 to 12 nanometers (see figure 7). This is a positive indication but it is not known if such an effect occurs in the submicron range or if the conductivity would increase more strongly for particles smaller than 12 nanometers. The study of size effects at fixed volume fraction is at the very center of our proposed study of transport of heat in nanoparticles.

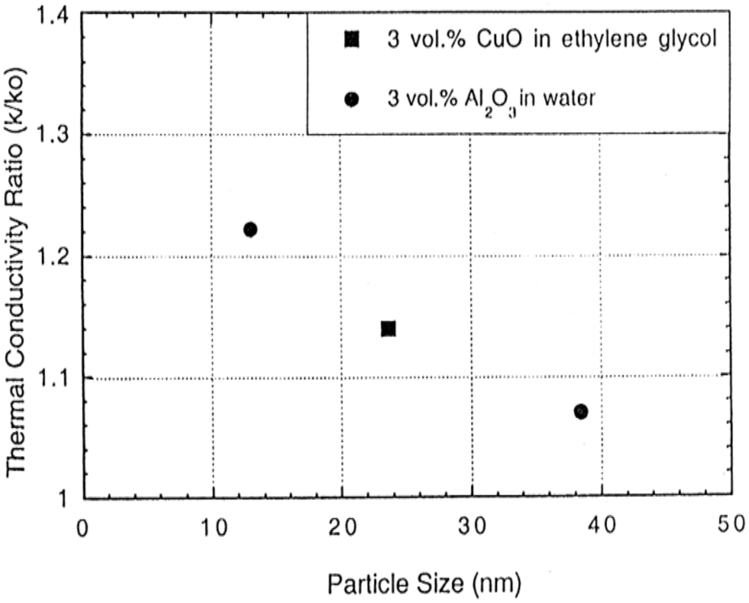

Figure 7. Thermal conductivity ratio of oxide nanofluids as a function of particle size at a fixed concentration (courtesy of S. Choi).

6 Conditions for advection of metallic nanoparticles

Whether or not the improved heat transport reported in the references on nanofluids are due to conduction or convection there is every reason to think that nanoparticles can markedly improve heat transport by convection.

Heat boats; advection of heat by nanoparticle. Metallic nanoparticles have a much higher heat conductivity than common liquids. For example, the thermal conductivity at room temperature of copper is 700 times greater than water and 3000 times greater than engine oil. However, the conductivity of some metallic nanoparticles can apparently decrease strongly at the smaller end of the nanoscale.[5] The large surface areas of these particles greatly promotes heat transport from the fluid to the particles in places where the fluid is hot and the release of heat from particles to fluid in places where the fluid is cold. To move heat by migration of nanoparticles they must be readily advected by the fluid.

Analogy to miscible liquids. Dispersion of nanoparticles in colloidal suspension look like miscible liquids with two different kinds of molecules, like glycerin and water. The nanoparticles are so small that they will follow the motion of the fluid; there is no need to consider the separate momentum of each phase. Instead the motion of nanoparticles could be described by diffusion, even using Fick’s law, as in miscible liquid. Nanofluids are different than most miscible liquids because the differences in the density of the two species can be much greater in nanofluids than in miscible liquids. This special property of huge weight differences of heavy metal particles in suspension should be studied.

7 Convection by concentration differences.

We are interested in convection of heat and our idea is that nanoparticles are a perfect vehicle for this transport. It is also true that the places occupied by nanoparticles in colloidal suspension are an important factor in convection. Consider copper particles with a density of 16 gm/cc in colloidal suspension in water. Let ϕ be the volume fraction of copper. The density of the copper-water mixture

$$\begin{aligned} \rho &= \rho_p\phi + \rho_w(1-\phi) = 16\phi + (1-\phi) \\ &= 15\phi + 1 \end{aligned} \tag{1}$$

If ϕ = 20%, ρ = 4 mainly due to copper. If by any fluid motion the copper is more concentrated in one place than another, very strong convection currents will develop under the action of the gravity term

$$[(\rho_p - \rho_w)\phi + \rho_w]g$$

in the Navier Stokes equation

$$\rho\frac{d\mathbf{u}}{dt} == \nabla p + [(\rho_p - \rho_w)\phi + \rho_w]g + \operatorname{div}(2\mu(\phi)\mathbf{D}[\mathbf{u}]) \tag{2}$$

where $\mathbf{D}[\mathbf{u}]$ is the rate of strain and $\mu(\phi)$ is the effective viscosity of the mixture. The basic motor for convection is from the gravity term; if $\phi(\mathbf{x},t)$ varies from place to place one part of the

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[5] A. Majumdar, 1998. Microscale Energy Transport in Solids, *Microscale Energy Transport*, C.L. Tien, A. Majumdar and F. Gerner, Eds., Taylor & Francis, Washington, DC.

mixture will rise or fall relative to another. In thermal convection the change in the density is induced by thermal expansion.

8 Boussinesq equations for nanofluids

In the Boussinesq equations[6] the density is related to the temperature by a linear equation of state

$$\rho - \rho_R = \alpha\theta, \quad \theta = T - T_R, \quad \alpha = \frac{d\rho}{dT_R}$$

where R is a reference state and α is the coefficient of thermal expansion. Here we expand ρ_p and ρ_w in the same way

$$\begin{aligned} \rho(\phi) &= (\rho_p - \rho_w)\phi + \rho_w \\ &= \tilde{\rho}(\phi) + \left[(\alpha_p - \alpha_w)\phi + \alpha_w\right]\theta \end{aligned} \tag{3}$$

where

$$\tilde{\rho}(\phi) = (\rho_{PR} - \rho_{WR})\phi + \rho_{WR} \ .$$

The difference $\alpha_p - \alpha_w$ of thermal expansivity of metal and water could have an important effect in driving convection governed by (4).

It may be convenient to introduce a constant ϕ_0 which is the nominal concentration of nanoparticles. Then $\delta\phi = \phi - \phi_0$,

$$\tilde{\rho}(\phi) = \tilde{\rho}(\phi_0) + (\rho_{PR} - \rho_{WR})\delta\phi \ .$$

After introducing

$$p = p_0 + \pi$$

we may remove $\tilde{\rho}(\phi_0)$ from the equations

$$-\nabla p_0 + \tilde{\rho}(\phi_0)\mathbf{g} = 0 \, .$$

The Boussinesq equations for our nanofluid is then

$$\rho_0 \frac{d\mathbf{u}}{dt} = -\nabla\pi + (\rho_{PR} - \rho_{WR})\delta\phi\,\mathbf{g} + \left[(\alpha_p - \alpha_w)\phi - \alpha_w\right]\theta\,\mathbf{g} + \operatorname{div}\left[\mu(\phi)\mathbf{D}[\mathbf{u}]\right]. \tag{4}$$

The equation expressing the conservation of mass is

$$\frac{d\rho(\phi)}{dt} + \rho(\phi)\ \operatorname{div}\mathbf{u} = 0 \tag{5}$$

and under the assumption that the deviation from the nominal concentration $\delta\phi$ is small this becomes

$$(\rho_{PR} - \rho_{WR})\frac{d\phi}{dt} + \tilde{\rho}(\phi_0)\ \operatorname{div}\mathbf{u} = 0 \tag{6}$$

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[6] D.D. Joseph, *Stability of Fluid Motions II*, Chap. VIII, Springer 1976

(We recall that in the usual Boussinesq equation div $\mathbf{u} = 0$).

Since our colloidal nanoparticles follow the liquid motion it is reasonable to regard this colloidal suspension of nanoparticles as a miscible mixture in which the separate motion is governed by a Fick's law leading to a diffusion equation

$$\frac{\partial \phi}{\partial t} + \mathbf{u} \cdot \nabla \phi = D(\phi) \nabla^2 \phi \tag{7}$$

with a to-be-determined nanodiffusion parameter D(ϕ).

Since we are interested in heat transport by advection of nanoparticles we introduce an appropriate Bousinesq energy equation

$$\rho C \left(\frac{\partial \theta}{\partial t} + \mathbf{u} \cdot \nabla \theta \right) = \text{div} \left(k(\phi) \nabla \theta \right) \tag{8}$$

In general all the coefficients $\rho(\phi)$, $\alpha(\phi)$, $\mu(\phi)$, $D(\phi)$, $C(\phi)$, $k(\phi)$, are functions of ϕ evaluated at the reference temperature. Probably these equations (4), (5), (7) and (8) should first be considered as constants evaluated at nominal concentration ϕ_0 of nanoparticles.

The Bousinesq equations for nanoparticles do not display an explicit dependence on a nanoparticle. The equations are meant to apply to a fluid mixture in which the particles follow the fluid; both with a velocity of **u**. This requires that the particles are colloidal which for heavy metallic particles probably implies that the particles are measured in nanometers. Colloidal particles can probably be modeled as miscible liquids in which the relative motion of the particles can be described by diffusion and the density of the mixture is heavily weighted by heavy particles.

9 Heat transport in phase change liquids

It is of interest to disperse nanoparticles in a liquid that can be solidified like molten and frozen glass, water and ice, and resins that have a liquid and solid phase. Heat transported by convection in the liquid phase can only be transported by conduction in the solid phase. If the increased heat transport is due to conduction we might see the same enhanced heat transport in the solid phase that we see in the liquid phase. There are many ways to create nanosolids from nanofluids by phase-change; even ice crystals laced with nanoparticles could be considered. We definitely plan to do these phase-change experiments but the details have not yet been worked out.

10 Microwave heating of nonpolar liquids

Holzwarth *et al* [1998][7] have shown that the microwave absorption characteristics of xylene as a model nonpolar solvent are dramatically increased by the incorporation of dispersed cobalt and magnetite nanoparticles. This result is very important. It opens a whole range of applications in many industries, particularly in the oil industry where viscosity reduction of heavy oils downhole and in fractured reservoirs and the promotion of certain chemical reactions are of

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[7] A. Holzwarth, J. Lou, T.A. Hatton and P.E. Laibinis, 1998. Enhanced microwave Heating of Nonpolar Solvents by Dispersed Magnetic Nanoparticles, *Ind. Eng. Chem. Res.*, **37**(7), 2701-2706.

particular interest. In previous applications of microwave heating the properties of polar molecules in water are essential.

Microwave energy penetrates a material and produces a volumetrically distributed heat source, due to molecular friction resulting from dipolar rotation of polar molecules and from the conductive migration of dissolved ions. The dipolar rotation is caused by variations of the magnetic and electrical field in the material. Heat is generated throughout the material, leading to faster heating rates and shorter processing times compared with conventional heating.

The results of Holzwarth *et al* show that this kind of volumetric microwave heating can be realized in nanofluids using appropriate nanoparticles. Magnetic particles offer superior microwave absorption characteristics over those of metals and polar liquids, accelerating the rate of microwave heating due to the effect of ferromagnetic resonance.

No literature is available where the combination of these elements is studied from the point of view of the convective heat transport in nanofluids and applied to nonpolar fluids like heavy viscous oil.

11 Proposed experiments

A key need in determining the enhanced value of nanofluids in many processes is gaining an understanding of the heat transfer characteristics under known conditions. These may then be compared to results with more common single-phase fluids under the same conditions. It has been shown that the addition of copper nanosized particles can have a significant impact on the bulk thermal conductivity as measured using standard techniques. The question arises whether the normal thermophysical properties of a nanofluid would translate directly into the normal heat transfer predictions and correlations used for single phase fluids.

A relatively simple heat transfer system consists of an enclosed fluid layer heated from one side and cooled from the other. In particular the classic case would be heated from below, Rayleigh-Bernard convection. In such a system, provided the aspect ratio is reasonably large, the time-average temperature distribution can be considered one-dimensional at sufficiently high Rayleigh number. No matter what the Rayleigh number, there are well-accepted correlations for the Nusselt number as a function of Rayleigh number. One proposed experiment would be a cell heated from below and cooled from above. Overall heat transfer measurement would be based on the electrical energy input to the bottom plate with a check heat balance from the energy taken out by water cooling the top surface. A recently designed apparatus was used here in an investigation of Rayleigh-Bernard natural convection at high Rayleigh numbers in gases under pressure, (Amy S. Fleischer, PhD Thesis, University of Minnesota, August 2000). A similar apparatus would be utilized for the proposed experiment in which the Nusselt number would be obtained as a function of Rayleigh number. In addition to the direct calorimetric measurements an interferometer would be used to study the temperature distribution in the fluid layer and serve as an additional quantitative check on the heat transfer and temperature measurements (see e.g. “Optical Systems for Flow Measurement: Shadowgraph, Schlieren, and Interferometric Techniques,” by R.J. Goldstein and T.H. Kuehn in *Fluid Mechanics Measurements*, edited by R.J. Goldstein, 1996).

In addition to natural convection studies a flow loop will be designed and constructed to compare forced convection heat transfer results for nanofluids. As a test of each heat transfer apparatus similar experiments will be done with single-phase fluids. Key parameters to vary in the experiments to study advection of heat by metallic nanoparticles include:

1) Composition of particles
2) Size and possible shape of particles
3) Particle concentration in fluid
4) Carrying fluid

One goal of the study is a correlation of the results with separately measured thermalphysical properties. Thus, in cooperation with Dr. Choi at Argonne National Laboratory, we would measure the thermal conductivity of the nanofluids using at least two different techniques, a transient vertical wire and a small cell heated from above. Other properties of the nanofluids, e.g., viscosity, bulk specific heat and density refraction would be determined.

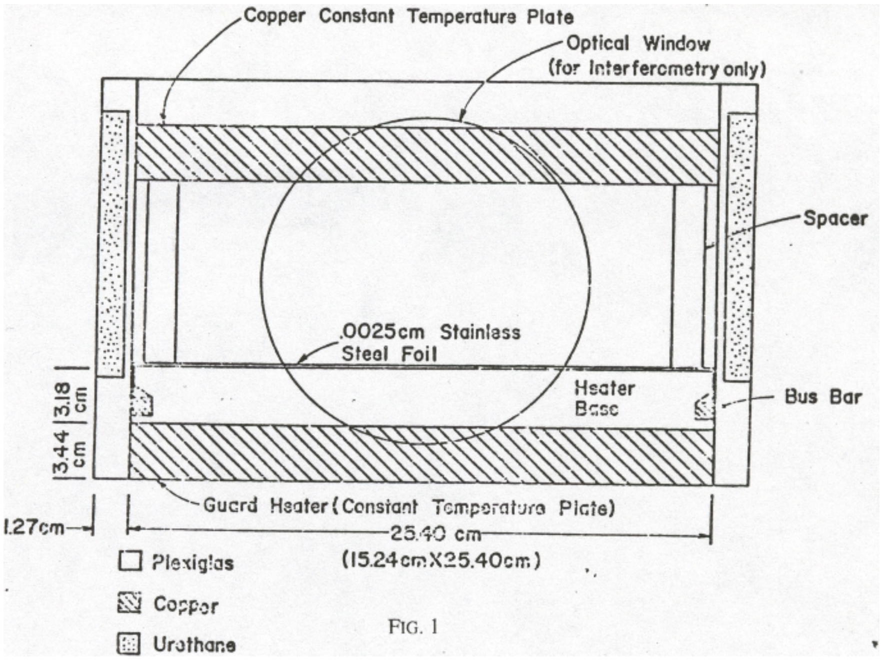

Figure 8.

12 Experiments on microwave irradiation of nanoparticles

The objective of the experiments is to understand and predict the behavior of nonpolar nanofluids and the heat transfer properties due to the irradiation of microwaves. In particular, the purpose of the experiments is to study the influence of parameters like the size, morphology and physical properties of the nanoparticle on the nanofluids dynamic; with the objective to understand the fluid transport properties of the highly viscous composite fluid.

Three types of nanoparticles that have been selected for the experiments: *magnetite*, *molybdenum* and *cobalt*. The size and dispersion of the particle diameters will be monitored. To prepare the dispersion, the particles will be coated with a surfactant that will remain active during the sampling preparation. The base fluid will be a highly viscous, transparent, non-mineral oil.

A microwave source of 900MHz and 2800MHz will be used. The direction of the irradiation will be controlled. The sample volumetric temperature profiles will be recorded after each irradiation. Due to microwave properties, special provisions are required; no metallic probe can be present during the irradiation process. There are ways to avoid metallic probes; one is to use small angle neutron scattering, which has been tested when high magnetic fields are present. Second, is to apply a pseudo continuous monitoring. The idea is to pump microwaves at intervals of 5 seconds and sample all the required data in the next 5 seconds.

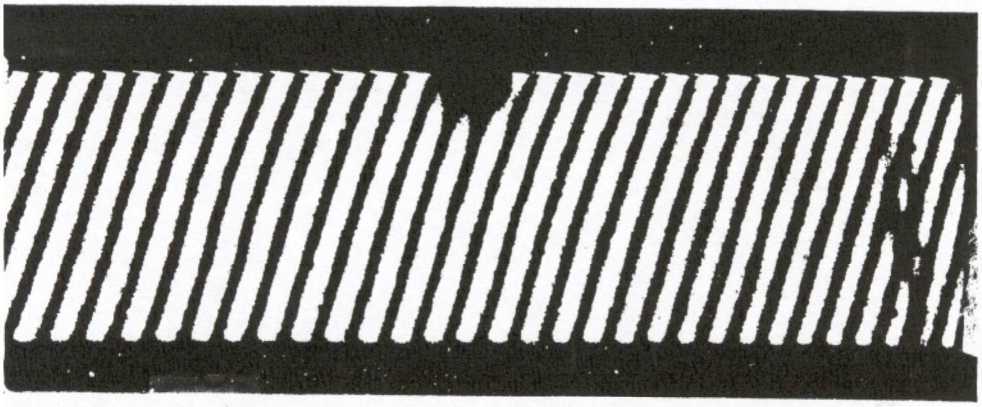

FIG. 2 "Quiescent Fluid" Vertical gap $\approx 1{\cdot}9$ cm; Rayleigh number $\approx 1{\cdot}69 \times 10^3$

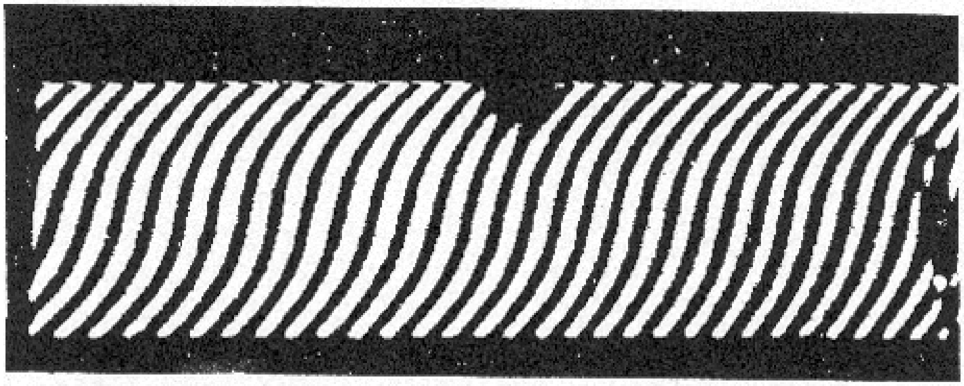

FIG. 3 "Cellular Flow" Vertical gap $\approx 1{\cdot}9$ cm; Rayleigh number $\approx 2{\cdot}84 \times 10^3$

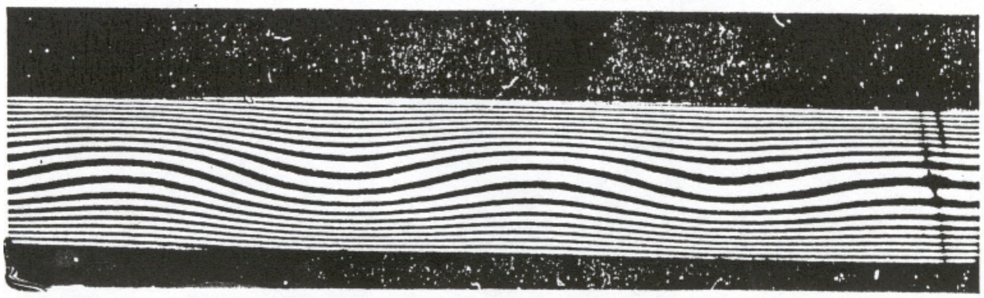

FIG. 4 "Cellular Flow" Vertical gap $\approx 1{\cdot}3$ cm; Rayleigh number $\approx 2{\cdot}63 \times 10^3$

Figure 9.

13 Size versus weight experiments

We seek the size of the particles of given density, which will not settle in a less dense liquid due to the action of Brownian motion. The simplest experiment is one of long duration in which the thickness of the sedimentation layer after weeks and months is monitored as a function of the size of the particles of fixed density. This might lead to a critical size statement within tolerance; say that we see sedimentation after one-month time.

The measurement of the concentration gradient is more difficult but can lead to an even more useful definition of the critical size of a particle. In this case we could look for a uniform dispersion over a prescribed distance in the sedimentation column. The larger the distance the more uniform the dispersion. This critical size could be ten or hundreds of nanometers for very heavy metallic and ferromagnetic (cobalt), particles. Knowledge of the critical size is an operational requirement for the preparation of nanofluids and it could be used as a scale length for a dimensionless description of the size. The concept of the gravitational length is useful as a guide for the critical size threshold, but the theory of gravitational length is not in good agreement with the experiments reports by Piazza *et al*, 1993. It is desirable to put up dedicated experiments to determine the critical size of heavy particles.

Several techniques are available for determining the concentration of particle suspended by Brownian forces. These techniques are all realizations for different conditions of dynamic light scattering (DLS[8]). A number of these realizations require that the particles and fluids have the same refractive index; this is a severe requirement. For dilute solutions index of refraction matching can be avoided using enhanced laser diffraction. Photon correlation spectroscopy (PCS), in which the time fluctuations of the intensity of light scattered by the particle dispersion are monitored, is widely used. Newly developed XPCS[9] techniques extend PCS methods to the x-ray region; obviously refractive index matching is not required for x-rays.

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[8] BJ Berne and R Pecora 2000. *Dynamic Light Scattering*, Dover Publications, New York.

[9] T. Thurn-Albrecht, et al 1999. *Phy Rev E*, 59, 642.