ENHANCING THERMAL CONDUCTIVITY OF FLUIDS WITH NANOPARTICLES*

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ABSTRACT

Low thermal conductivity is a primary limitation in the development of energy-efficient heat transfer fluids that are required in many industrial applications. In this paper we propose that an innovative new class of heat transfer fluids can be engineered by suspending metallic nanoparticles in conventional heat transfer fluids. The resulting "nanofluids" are expected to exhibit high thermal conductivities compared to those of currently used heat transfer fluids, and they represent the best hope for enhancement of heat transfer. The results of a theoretical study of the thermal conductivity of nanofluids with copper nanophase materials are presented, the potential benefits of the fluids are estimated, and it is shown that one of the benefits of nanofluids will be dramatic reductions in heat exchanger pumping power.

NOMENCLATURE

d	Pipe diameter
f	Fanning friction factor
h	Heat transfer coefficient
k	Thermal conductivity
L	Length
n	Shape factor
Nu	Nusselt number
P	Pumping power
Pr	Prandtl number
Re	Reynolds number
v	Velocity
α	Particle volume fraction
δp	Pressure drop
ρ	Density
ψ	Sphericity

Subscripts

eff	Effective
m	Metallic particle
nf	Nanofluid
o	Reference fluid without nanoparticles

INTRODUCTION

Fluids are often used as heat carriers in heat transfer equipment. Examples of important uses of heat transfer fluids include vehicular and avionics cooling systems in the transportation industry, hydronic heating and cooling systems in buildings, and industrial process heating and cooling systems in petrochemical, textile, pulp and paper, chemical, food, and other processing plants. In all of these applications, the thermal conductivity of heat transfer fluids plays a vital role in the development of energy-efficient heat transfer equipment. With an increasing global competition, industries have a strong need to develop advanced heat transfer fluids with significantly higher thermal conductivities than are presently available.

Despite considerable previous research and development efforts on heat transfer enhancement, major improvements in cooling capabilities have been constrained because of the low thermal conductivity of conventional heat transfer fluids. However, it is well known that at room temperature, metals in solid form have orders-of-magnitude larger thermal conductivities than fluids. For example, the thermal conductivity of copper at room temperature is ≈ 700 times greater than that of water and ≈ 3000 times greater than that of engine oil, as shown in Table 1. The thermal conductivity of metallic liquids is much greater than that of nonmetallic liquids. Therefore, the thermal conductivities of fluids that contain suspended solid metallic particles are expected to be significantly enhanced when compared with conventional

Table 1. THERMAL CONDUCTIVITY (**W/m-K**) OF VARIOUS MATERIALS AT 300 K UNLESS OTHERWISE NOTED

Material	Thermal Conductivity
Metallic Solids	
Silver	429
Copper	401
Aluminum	237
Nonmetallic Solids	
Silicon	148
Metallic Liquids	
Sodium @ 644 K	72.3
Nonmetallic Liquids	
Water	0.613
Engine oil	0.145

heat transfer fluids. In fact, numerous theoretical and experimental studies of the effective thermal conductivity of dispersions that contain solid particles have been conducted since Maxwell's theoretical work was published more than 100 years ago (Maxwell, 1881). However, all of the studies on thermal conductivity of suspensions have been confined to millimeter- or micrometer-sized particles. Maxwell's model shows that the effective thermal conductivity of suspensions that contain spherical particles increases with the volume fraction of the solid particles. It is also known that the thermal conductivity of suspensions increases with the ratio of the surface area to volume of the particle.

It is proposed that nanometer-sized metallic particles can be suspended in industrial heat transfer fluids such as water, ethylene glycol, or engine oil to produce a new class of engineered fluids with high thermal conductivity. The author has coined the term nanofluids (NFs) for this new class of engineered heat transfer fluids, which contain metallic particles with average particle sizes of about 10 nanometers and can be produced by current nanophase technology.

Nanofluids are expected to exhibit superior properties when compared with conventional heat transfer fluids and fluids that contain micrometer-sized metallic particles. Because heat transfer takes place at the surface of the particle, it is desirable to use a particle with a large surface area. Nanoparticles have extremely large surface areas and therefore have a great potential for application in heat transfer. The much larger relative surface areas of nanophase powders, when compared with conventional micrometer-sized powders, should markedly improve the heat transfer capabilities and stability of the suspensions.

Researchers at Argonne National Laboratory (ANL) have been developing advanced fluids for industrial applications, including district heating and cooling systems (Choi and Tran, 1991; Choi et al., 1992a and 1992b). One of the problems identified in this R&D program was that micrometer-sized particles cannot be used in practical heat transfer equipment because of severe clogging problems. However, nanophase metals are believed to be ideally suited for applications in which fluids flow through small passages, because the metallic nanoparticles are small enough that they are expected to behave like molecules of liquid. Therefore, nanometer-sized particles will not clog flow passages, but will improve the thermal conductivity of the fluids. This will open up the possibility of using nanoparticles even in microchannels for many envisioned high-heat-load applications. More recently, a project was begun at ANL to demonstrate the feasibility of the concept of nanofluids. Successful employment of nanofluids will result in significant energy and cost savings and will support the current industrial trend towards component miniaturization by enabling the design of smaller and lighter heat exchanger systems.

The purpose of the paper is to demonstrate theoretically the feasibility of the concept of nanofluids. After briefly describing the technology for producing nanoparticles and suspensions, we shall estimate the thermal conductivity of nanofluids with copper nanophase materials and the subsequent heat transfer enhancement as a function of thermal conductivity. We will also explore the potential benefits of nanofluids in the expectation that the ultra-high-performance nanofluids may have major implications for many industries.

TECHNOLOGY FOR PRODUCTION OF NANOPARTICLES AND SUSPENSIONS

Modern fabrication technology provides great opportunities to actively process materials on micro- and nanometer scales. Materials with novel properties can be produced on nanometer scales. Nanostructured or nanophase materials are nanometer-sized solid substances engineered on the atomic or molecular scale to produce either new or enhanced physical properties not exhibited by conventional bulk solids. All physical mechanisms have a critical length scale, below which the physical properties of materials are changed. Therefore, particles < 100 nm in diameter exhibit properties different from those of conventional solids. The noble properties of nanophase materials come from the relatively high surface-area-to-volume ratio that is due to the high proportion of constituent atoms that reside at the grain boundaries. The thermal, mechanical, optical, magnetic, and electrical properties of nanophase materials are superior to those of conventional materials with coarse grain structures. Consequently, the exploration in research and development of nanophase materials has drawn considerable attention from material scientists and engineers alike (Duncan and Rouvray, 1989; Siegel, 1991).

Much progress has been made in the production of nanophase materials, and current nanophase technology can produce large quantities of powders with average particle sizes in the 10-nm range. Several "modern" nanophase materials have been prepared by physical gas-phase condensation or chemical synthesis techniques (Siegel, 1991). The gas-phase condensation process involves the evaporation of a source material and the rapid condensation of vapor into nanometer-sized crystallites or loosely agglomerated clusters in a cool, inert, reduced-pressure atmosphere. A chemistry-based

solution-spray conversion process starts with water-soluble salts of source materials. The solution is then turned into an aerosol and dried by a spray-drying system. Rapid vaporization of the solvent and rapid precipitation of the solute keeps the composition identical to that of the starting solution. The precursor powder is then placed in a fluidized-bed reactor to evenly pyrolyze the mixture, drive off volatile constituents, and yield porous powders with a uniform homogeneous fine structure (Ashly, 1994). A third technique is to generate nanophase materials by condensation of metal vapors during rapid expansion in a supersonic nozzle (Hill, et al., 1963; Andres, et al., 1981; Brown, et al., 1992).

If powders are produced by one of these processes, some agglomeration of individual particles may occur. It is well known, however, that these agglomerates, which are typically 1 micrometer or so in size, require little energy to fracture into smaller constituents, and thus it is possible they will not present a problem in this application. If, however, agglomeration is a problem, it would prevent realization of the full potential of high surface areas of nanoparticles in nanofluids. Under such conditions, these conventional technologies for production of nanophase materials are not suitable for nanofluids.

Another promising technique for producing nonagglomerating nanoparticles involves condensing nanophase powders from the vapor phase directly into a flowing low vapor pressure fluid. This technique was developed in Japan more than 10 years ago by Akoh et al. (1978), but has been essentially ignored by the nanocrystalline-materials community because of difficulties in subsequently separating the particles that are produced from the fluids to make dry powders or bulk materials by sintering individual nanometer-sized particles.

THEORETICAL STUDY OF THERMAL CONDUCTIVITY OF NANOFLUIDS

Because of the absence of a theory for the thermal conductivity of nanofluids, two existing models that were developed for conventional solid-liquid systems with fine particles are used in this study to estimate the effective thermal conductivity of nanofluids. Batchelor and O'Brien (1977) have developed an expression for the effective thermal conductivity k_{eff}, which is applicable to two-phase systems that contain metal powders with particle diameters on the order of micrometers, i. e.,

$$k_{eff}/k_0 = 4 \ln(k_m/k_0) - 11. \tag{1}$$

where k_m is the thermal conductivity of the metallic particle and k_0 is the thermal conductivity of the reference fluid. However, it should be noted that the theory of Batchelor and O'Brien (1977) was originally developed for a point-contact porous medium. When there is no contact between the particles, the effective thermal conductivity is independent of the conductivity ratio. Thus, for values of the conductivity ratio ranging from 100 to 10,000, the effective thermal

conductivity of noncontacting systems is estimated from the equation

$$k_{eff}/k_0 = 4 \tag{2}$$

If it is assumed that this expression is applicable to nanofluids, nanoparticles are expected to increase the thermal conductivity of the base fluids by a factor of 4. However, this expression seems unfeasible for nanofluids because it does not involve the particle volume fraction or particle shape.

Hamilton and Crosser (1962) have developed a more elaborate model for the effective thermal conductivity of two-component mixtures as a function of the conductivity of the pure materials, the composition of the mixture, and the shape of the dispersed particles. For mixtures in which the ratio of conductivities of two phases is > 100, the effective thermal conductivity of two-component mixtures can be calculated as follows:

$$k_{eff}/k_0 = [k_m+ (n-1) k_0 - (n-1) \alpha (k_0 - k_m)] / [k_m+ (n-1) k_0 + \alpha (k_0 - k_m)] \tag{3}$$

where α is the particle volume fraction and n is the empirical shape factor given by

$$n = 3 / \psi, \tag{4}$$

where ψ is the sphericity, defined as the ratio of the surface area of a sphere with a volume equal to that of the particle to the surface area of the particle. This model shows that nonspherical shapes (all other circumstances being the same) will increase the conductivity above that of spheres.

Applying the Hamilton and Crosser model to copper nanoparticles in water, the effective thermal conductivity of the copper-water system has been estimated for three values for ψ. The effects of particle volume fraction and sphericity on the thermal-conductivity ratio for a copper-water system are plotted in Fig. 1. The results clearly show that the thermal conductivity of the fluid-particle system depends on both the particle volume fraction and the shape. Assuming that the sphericity of copper nanoparticles is 0.3, the thermal conductivity of water can be enhanced by a factor of 1.5 at the low volume fraction of 5% and by a factor of almost 3.5 at the high volume fraction of 20%. This finding demonstrates, theoretically, the feasibility of the concept of nanofluids, i.e., metallic nanoparticles are capable of significantly increasing the thermal conductivity of conventional heat transfer fluids. Furthermore, Masuda et al. (1993) have shown experimentally that γ-Al_2O_3 particles at a volume fraction of 4.3% can increase the effective thermal conductivity of water by $\approx 30\%$. The agreement between the estimated and measured conductivities is satisfactory.

POTENTIAL BENEFITS OF NANOFLUIDS

For turbulent convection transfer of heat in smooth pipes, the heat transfer coefficient can be calculated from the Dittus-Boelter correlation,

$$Nu = 0.023 \, Re^{0.8} \, Pr^{1/3}. \tag{5}$$

If it is assumed that only the thermal conductivity of the nanofluid system varies and other properties, such as the specific heat, density, and dynamic viscosity, are the same as for the reference fluid, then we obtain from Eq. 5,

$$h \sim v^{0.8} \, k^{2/3}, \tag{6}$$

which shows that the heat transfer coefficient h may be increased by increasing the velocity v or the thermal conductivity of the fluid k.

In heat exchangers that use conventional fluids, the heat transfer coefficient may only be increased by significantly increasing the velocity of the fluid in the heat transfer equipment. However, the pumping power significantly increases with increasing velocity. The frictional pressure drop for fully developed turbulent flows in a pipe is given as

$$\delta p = 2f\rho L \, v^2/d, \tag{7}$$

where ρ is the density of the fluid, L the length of the pipe, d the pipe diameter, and f the Fanning friction factor given by

$$f = 0.079 \, Re^{-0.25}. \tag{8}$$

It can be shown that the frictional pressure drop is given by the relationship

$$\delta p \sim v^{1.75}. \tag{9}$$

Because pumping power P is proportional to the product of the pressure drop and the flow rate, it can be expressed by the relationship

$$P \sim v^{2.75}. \tag{10}$$

From Eqs. 6 and 10, enhancement of heat transfer due to increased pumping power can be estimated from the following equation:

$$h/h_o = (P/P_o)^{0.29}. \tag{11}$$

For a nanofluid flowing in the same heat transfer equipment at a fixed velocity, enhancement of heat transfer due to increased thermal conductivity can be estimated from the equation

$$h_{nf}/h_o = (k_{nf}/k_o)^{2/3}. \tag{12}$$

The effects of thermal conductivity and pumping power on heat transfer are plotted in Fig. 2. In heat exchangers that use conventional fluids, heat transfer can only be improved by significantly increasing flow rates. For example, to improve the heat transfer by a factor of 2, the pumping power should be increased by a factor of ≈ 10. However, if a nanoparticle-based fluid with a thermal conductivity of ≈ 3 times that of a conventional fluid were used in the same heat transfer equipment, the rate of heat transfer would be doubled.

Liu et al. (1988) have studied the influence of particle loading and size on the pressure drop of slurry. Their data show that solids suspensions in the 20% volume fraction range incur little or no penalty in pressure drop as compared with single-phase fluids of comparable flow rate. Therefore, it is reasonable to assume that the nanofluid pressure drop behaves like that of a single-phase fluid at volume fractions up to 20%. Then, the potential savings in pumping power is particularly significant as the heat transfer enhancement ratio is increased, as shown in Fig. 3. This could lead to a major technological breakthrough in the development of energy-efficient industrial heat transfer fluids. Therefore, the potential benefits of nanofluids could provide tremendous performance, size/weight, and cost advantages.

FUTURE RESEARCH PLANS

The research effort to produce and characterize the heat transfer behavior of nanofluids will consist of five main tasks.

1. Nanophase metal powders will be produced in existing state-of-the-art gas-condensation preparation systems at ANL. The particle size and agglomeration behavior of nanophase powders in liquids will be studied.

2. A new technique for producing nonagglomerating nanoparticles for nanofluids by directly condensing nanophase powders into a flowing fluid will be developed, based on the system designed by Akoh et al. (1978). The properties of nanofluids produced by this technique will be compared with those produced by inert-gas condensation.

3. Technology for production of nanoparticle suspensions will be developed and the stability, dispersion, and rheological/transport properties of these nanofluids will be investigated.

4. The flow characteristics of dilute and concentrated suspensions of nanoparticles will be studied. Heat transfer tests with nanoparticles in a range of up to 10 volume fraction will be conducted to demonstrate the expected dramatic improvement in energy efficiency from nanofluids.

5. Practical applications of nanofluids will be investigated.

CONCLUDING REMARKS

The concept of nanofluids is an innovative idea. The feasibility of the concept of high-thermal-conductivity nanofluids has been demonstrated by applying the Hamilton and Crosser (1962) model to copper nanoparticles in water, together with some experimental results of Masuda, et al. (1993) for γ-Al_2O_3 particles in water. The potential benefits of nanofluids with copper nanophase materials have been estimated. One of the benefits of nanofluids will be dramatic

reductions in heat exchanger pumping power. For example, to improve the heat transfer by a factor of 2, the pumping power with conventional fluids should be increased by a factor of ≈ 10. However, if a nanoparticle-based fluid with a thermal conductivity of ≈ 3 times that of a conventional fluid were used in the same heat transfer equipment, the nanoparticle-based fluid would double the rate of heat transfer without an increase in pumping power. The invention of nanofluids presents new challenges and opportunities for thermal scientists and engineers.

ACKNOWLEDGMENTS

REFERENCES

Akoh, H., Tsukasaki, Y., Yatsuya, S., and Tasaki, A., 1978, "Magnetic Properties of Ferromagnetic Ultrafine Particles Prepared by a Vacuum Evaporation on Running Oil Substrate," *J. Cryst. Growth*, 45, pp. 495-500.

Andres, R. P., Bowles, R. S., Kolstad, J. J., and Calo, J. M., 1981, "Generation of Molecular Clusters of Controlled Size," *Surface Sci.*, 106, pp. 117-124.

Ashly, S., 1994, "Small-scale Structure Yields Big Property Payoffs," *Mechanical Engineering*, Vol. 116, No. 2, pp. 52-57.

Batchelor, G. K. and O'Brien, R. W., 1977, "Thermal or Electrical Conduction through a Granular Material," *Proc. R. Soc. Lond.*, A355, pp. 313-333.

Brown, D. P., Chung, J. N., and Crowe, C. T., 1992, "A Numerical Simulation of Nanocluster Formation in Supersonic Expansion Flows," *Micromechanical Systems*, ASME DSC-Vol. 40, pp. 211-225.

Choi, U. S., Cho Y. I., and Kasza, K. E.. 1992a, "Degradation Effects of Dilute Polymer Solutions on Turbulent Friction and Heat Transfer Behavior," *J. Non-Newtonian Fluid Mechanics*, 41, pp. 289-307.

Choi, U. S., France, D. M., and Knodel, B. D., 1992b, "Impact of Advanced Fluids on Costs of District Cooling Systems," *Proc. 83rd Ann. Int. District Heating and Cooling Assoc. Conf.*, Danvers, MA, June 13-17., The Int. District Heating and Cooling Assoc., Washington, D.C., pp. 343-359.

Choi, U. S. and Tran, T. N., 1991, "Experimental Studies of the Effects of Non-Newtonian Surfactant Solutions on the Performance of a Shell-and-Tube Heat Exchanger," *Recent Developments in Non-Newtonian Flows and Industrial Applications*, eds. D. A. Siginer and M. N. Dhaubhadel, The American Society of Mechanical Engineers, New York, FED–Vol. 124, pp. 47-52.

Duncan, M. A. and Rouvray, D. H., 1989, "Microclusters," *Scientific American*, Dec., pp. 110-115.

Hamilton, R. L. and Crosser, O. K., 1962, "Thermal Conductivity of Heterogeneous Two-Component Systems," *I & EC Fundamentals*, Vol. 1, No. 3, pp. 187-191.

Hill, P. G., Witting, H., and Demetri, E. P., 1963, "Condensation of Metal Vapors During Rapid Expansion," *J. Heat Transfer*, Nov., pp. 303-317.

Liu, K. V., Choi, U. S., and Kasza, K. E., 1988, "Measurements of Pressure Drop and Heat Transfer in Turbulent Pipe Flows of Particulate Slurries," Argonne National Laboratory Report, ANL-88-15.

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 γ-Al_2O_3, SiO_2, and TiO_2 Ultra-fine Particles)," *Netsu Bussei(Japan)*, Vol. 4, No. 4, pp. 227-233.

Maxwell, J. C., 1881, *A Treatise on Electricity and Magnetism*, 2nd ed., 1, 435, Clarendon Press.

Siegel, R. W., 1991, "Cluster-Assembled Nanophase Materials," *Annual Review of Materials Science*, 21, pp. 559-578.
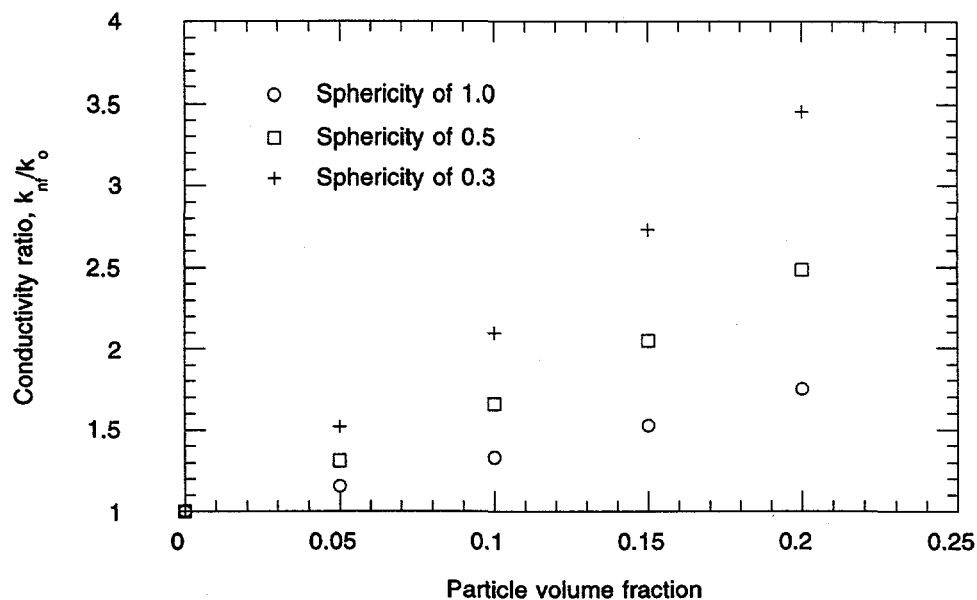
Figure 1. EFFECT OF PARTICLE VOLUME FRACTION AND SPHERICITY
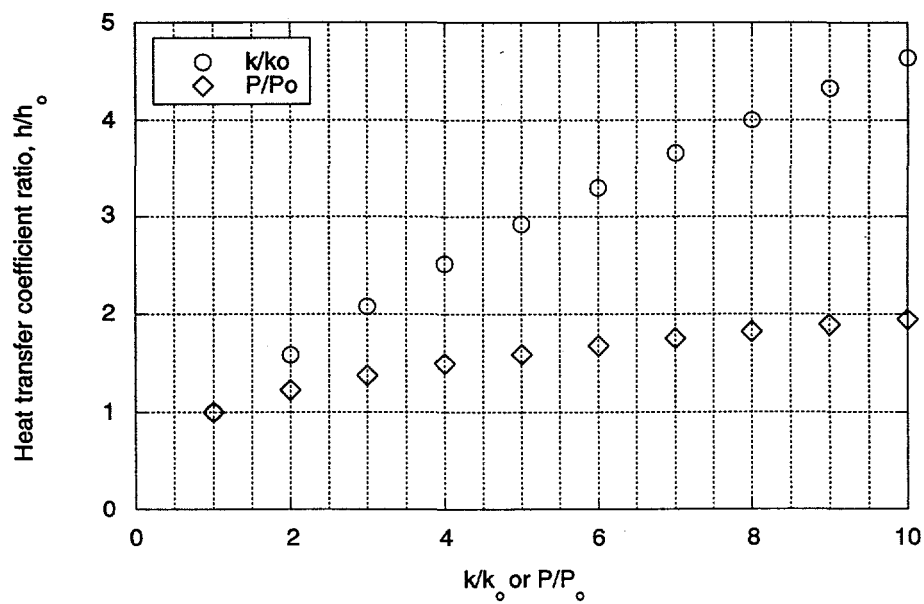ON THERMAL CONDUCTIVITY RATIO FOR COPPER-WATER SYSTEM

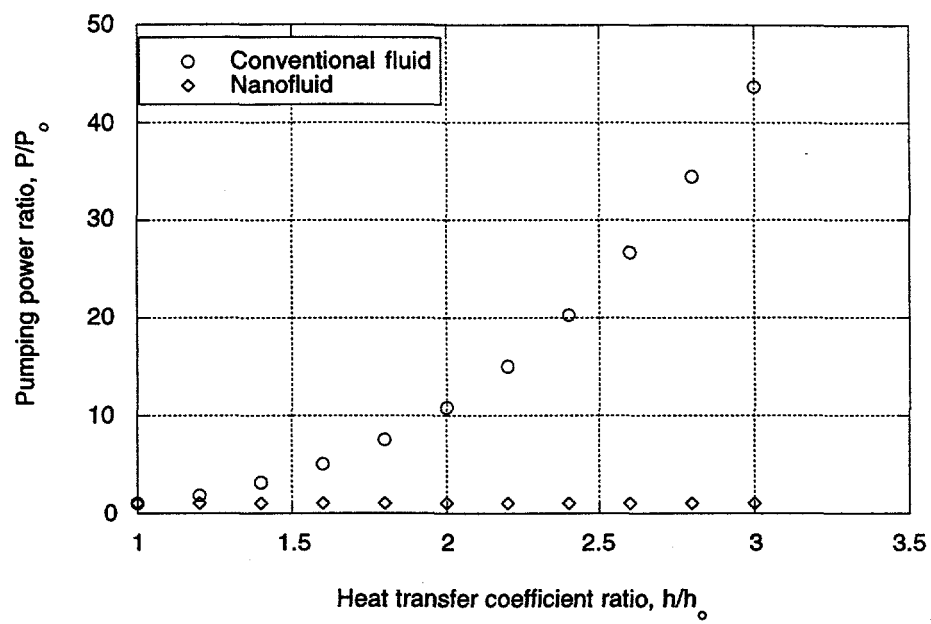Figure 2. EFFECTS OF THERMAL CONDUCTIVITY AND PUMPING POWER
ON HEAT TRANSFER

Figure 3. POTENTIAL PUMPING POWER SAVINGS WITH NANOFLUIDS