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Nanofluids for Heat Transfer: An Engineering Approach

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Abstract

An overview of systematic studies that address the complexity of nanofluid systems and advance the understanding of nanoscale contributions to viscosity, thermal conductivity, and cooling efficiency of nanofluids is presented. A nanoparticle suspension is considered as a three-phase system including the solid phase (nanoparticles), the liquid phase (fluid media), and the interfacial phase, which contributes significantly to the system properties because of their extremely high surface-to-volume ratio in nanofluids. The systems engineering approach was applied to nanofluid design resulting in a critical assessment of various parameters in the

multivariable nanofluid systems. Understanding the relative importance of nanofluid parameters for heat transfer allows engineering nanofluids with desired set of properties.

Keywords: nanofluid, systems engineering, heat transfer, efficiency, nanoparticle suspension, particle size, particle shape, base fluid, interfacial thermal resistance.

Introduction

Suspensions of solid submicron and nanometer-sized particles in various fluids (nanofluids) have been considered for applications as advanced heat transfer fluids for almost two decades. However due to the wide variety and the complexity of the nanofluid systems, no agreement has been achieved on the magnitude of potential benefits of using nanofluids for heat transfer applications. Large volume of studies devoted to characterization of individual thermo-physical properties of nanofluids, such as thermal conductivity, viscosity, and agglomeration of nanoparticles has been summarized in a number of review articles [1-9].

Evaluation of cooling efficiency, i.e. ability to remove heat from the heat source, includes assessing contributions from thermal conductivity, viscosity, specific heat, and density of the fluid and also depends on the applied flow regime. The studies devoted to evaluation of the heat transfer performance of nanofluids are scarce and inconclusive compared to the studies on the thermo-physical properties of various nanofluids indicating a significant gap between fundamental research and practical applications of nanofluids for thermal management.

In this paper we present a summary of systematic experimental studies of both thermo-physical properties and heat transfer in nanofluids. We believe that the underestimated complexity and the controversy of nanofluid systems is related to the solid/liquid boundary

layers between nanoparticles and the liquid, which at significant surface area of nanoparticles contribute to the fluid properties, resulting in three-phase systems. The approach to nanofluids as three-phase systems (instead of traditional consideration of nanofluids as two-phase systems of solid and liquid) allows for deeper understanding of correlations between the engineering parameters, nanofluid properties, and cooling performance. The factors contributing to the fluid cooling efficiency are discussed first, followed by a review of nanofluid engineering parameters and a brief analysis of their contributions to basic thermo-physical properties. Finally, an engineering approach is used to describe how various nanofluid parameters contribute to the systems cooling performance. The latter also offers insights into the principles of the efficient nanofluid design.

I. Cooling efficiency of nanofluids

The initial promise of nanofluids as advanced heat transfer fluids was based on the increased thermal conductivity of nanoparticle suspensions. Low thermal conductivity of conventional fluids improves when the solid particles are added. However the magnitudes of the effects reported in the literature are scattered from few percent (as predicted by effective medium theory (EMT) [10-12]) to hundred percents per volume fraction of nanoparticles (i.e. abnormal enhancements [4, 13-14]). Theoretical works exploring the mechanisms that could be responsible for abnormally enhanced thermal conductivities are widely presented in the literature [2, 15].

Unfortunately it is not always realized that the thermal conductivity is not the only property that determines the efficiency of heat transfer in the system. In the forced flow systems the coolant is pumped through the pipes of a heat exchanger, introducing convective heat transfer mechanisms and pumping power penalties. Efficiencies of various liquid coolants depend on the

fluid properties and the flow mode (laminar or turbulent) and can be estimated from the fluid dynamics equations [16]. In the case of fully developed laminar flow, the heat transfer coefficient (h) is proportional to the thermal conductivity (k), and independent of the flow velocity (within the acceptable range of inlet/outlet temperature difference) [17]:

$$h \propto k \qquad (1).$$

An alternative merit criterion for laminar flow [18] was suggested, for situation, when the tube diameter can be increased for the nanofluid to result in the same heat transfer coefficient:

$$\frac{\mu_{eff}}{\mu_0} \approx 1 + C_\mu \phi \; ; \quad \frac{k_{eff}}{k_0} \approx 1 + C_k \phi \; ; \quad C_\mu / C_k < 4 \qquad (2),$$

where ϕ is the particle volume fraction, μ is the dynamic viscosity of the nanofluid (*eff*) and the base fluid (*0*), and C_μ and C_k are viscosity and thermal conductivity enhancement coefficients, determined from experimental viscosity and thermal conductivity ratios. However it is not very practical when efficiencies of two fluids are compared in the same system geometry (i.e. tube diameter).

In turbulent flow regime the heat transfer rate (based on the Dittus-Boelter equation for heating applications) is dependent not only upon the thermal conductivity (k), but also on the density (ρ), specific heat (c_p), viscosity (μ) and flow velocity (V) [16]:

$$h \propto \rho^{4/5} c_p^{2/5} \mu^{-2/5} k^{3/5} V^{4/5} \qquad (3).$$

Introduction of nanoparticles to the fluid affects all of thermo-physical properties and should be accounted for in the nanofluid evaluations [18-19]. Density and specific heat are proportional to the volume ratio of solid and liquid in the system, generally with density increasing and specific heat decreasing with addition of nanoparticles to the fluid. The increase

in the thermal conductivity of nanofluids favors the heat transfer coefficient; however the well described increase in the viscosity of nanoparticle suspensions is not beneficial for heat transfer. The velocity term in the equation (3) represents the pumping power penalties resulting from the increased viscosity of nanofluids.

For comparing two liquid coolants flowing over or through a given geometry at a fixed velocity the ratio of Mouromtseff values (Mo) were suggested as a figure of merit [20-21]. The fluid with the highest Mo value will provide the highest heat transfer rate for cooling application, by the Dittus-Boelter equation:

$$Mo = \frac{\rho^{0.8} k^{0.67} c_p^{0.33}}{\mu^{0.47}} \; ; \; Mo_{eff} / Mo_0 > 1 \qquad (4).$$

Thus, the challenge in the development of nanofluids for heat transfer applications is in understanding of how micro- and macroscale interactions between the particles and the fluid affect the properties of the fluid. This requires a complex approach that accounts for changes in all important thermo-physical properties caused by introduction of nanomaterials to the fluid. It is obvious that the properties of suspensions depend on many system variables (i.e. engineering parameters) such as the material, concentration, size, and shape of the nanoparticles, the properties of the base fluid, and the presence of additives, surfactants, electrolyte strength, and pH. Below we discuss how each of the above parameters affects individual nanofluids properties.

II. Nanofluid Engineering Parameters

a. Nanoparticles

Great varieties of nanoparticles are commercially available and can be used for preparation of nanofluids. Nanoparticle material, concentration, size and shape all contribute to the nanofluid properties.

Nanoparticle material defines density, specific heat and thermal conductivity of the solid phase contributing to nanofluids properties (subscripts *p*, *0,* and *eff* refer to nanoparticle, base fluid and nanofluid respectively) in proportion to the volume concentration of particles (ϕ):

$$\rho_{eff} = (1-\phi)\rho_0 + \phi\rho_p \quad (5);$$

$$(c_p)_{eff} = \frac{(1-\phi)(\rho c_p)_o + \phi(\rho c_p)_p}{(1-\phi)\rho_0 + \phi\rho_p} \quad (6);$$

$$k_{eff} = k_0\left(\frac{k_p + 2k_0 + 2(k_p - k_0)\phi}{k_p + 2k_0 - (k_p - k_0)\phi}\right)$$

, (for the simplest case of spherical particles by EMT) (7).

As it was mentioned previously the materials with the higher thermal conductivity, specific heat, and density are beneficial for heat transfer.

The size of nanoparticles defines the surface-to-volume ratio and for the same volume concentrations suspension of smaller particles will have a higher area of the solid/liquid interface. Therefore the contribution of interfacial effects will be stronger in such a suspension [22-23]. Interactions between the nanoparticles and the fluid are manifested through the interfacial thermal resistance, also known as Kapitza resistance (R_k), that arises because interfaces act as an obstacle to heat flow and diminish the overall thermal conductivity of the system [24]. The values of Kapitza resistance are constant for the particular solid/liquid interface defined by the strength of solid-liquid interaction and were correlated to the wetting properties of the interface [24]. When the interactions between the nanoparticle surfaces and the fluid are weak (non-wetting case) the rates of energy transfer are small resulting in relatively large values

of R_k. The overall negative contribution of the solid/liquid interface to the macroscopic thermal conductivity of nanofluids was found proportional to the total area of the interface, increasing with decreasing particle sizes [23, 25].

The size of nanoparticles also affects the viscosity of nanofluids. Generally viscosity increases as the volume concentration of particles increases. Studies of suspensions with the same volume concentration and material of nanoparticles but different sizes [25-26] showed that the viscosity of suspension increases as the particle size decreases. This behavior is related to formation of structured layers of fluid along the nanoparticle interfaces that move with the particles in the flow [27]. The thicknesses of those fluid layers depend on the strength of particle-fluid interactions while the volume of immobilized fluid increases in proportion to the total area of the solid/liquid interface. The "effective volume concentration" (immobile fluid and nanoparticles) is higher in suspensions of smaller nanoparticles resulting in higher viscosity. Therefore contributions of interfacial effects, negligible at micron particle sizes become very important for nanoparticle suspensions. To achieve higher thermal conductivity and lower viscosity beneficial for heat transfer the suspensions of larger nanoparticles should be used.

A drawback of using larger nanoparticles is the potential instability of nanofluids. Rough estimation of the settling velocity of nanoparticles (V_s) can be calculated from Stokes law (only accounts for gravitational and buoyant forces):

$$V_s = \frac{2}{9}\left(\frac{\rho_p - \rho_0}{\mu}\right) r^2 g \qquad (8),$$

where g is the gravitational acceleration. As one can see from the equation (8), the stability of a suspension (defined by lower settling rates) improves if: (a) the density of the solid material (ρ_p)

is close to that of the fluid (ρ_0); (b) the viscosity of the suspension (μ) is high; (c) the particle radius (r) is small.

Effects of the nanoparticles shapes on the thermal conductivity and viscosity of alumina-EG/H_2O suspensions [23] are also strongly related to the total area of the solid/liquid interface. In nanofluids with non-spherical particles the thermal conductivity enhancements predicted by the Hamilton-Crosser equation [11] (randomly arranged elongated particles provide higher thermal conductivities than spheres, EMT [28]) are diminished by the negative contribution of the interfacial thermal resistance as the sphericity of nanoparticles decreases. Elongated particles and agglomerates also result in higher viscosity at the same volume fraction as spheres due to structural limitation of rotational and transitional Brownian motion. Therefore it can be concluded that spherical particles or low aspect ratio spheroids are more practical for achieving lower viscosities in nanofluids – the property that is highly desirable for minimizing the pumping power penalties in cooling system applications.

b. Base fluid

The influence of base fluids on the thermo-physical properties of suspensions is not very well studied and understood. However there are few publications indicating some general trends of the base fluid effects.

Suspensions of the same Al_2O_3 nanoparticles in water, ethylene glycol (EG), glycerol, and pump oil showed increase in relative thermal conductivity (k_{eff}/k_0) with decrease in thermal conductivity of the base fluid [22, 29]. On the other hand the alteration of the base fluid viscosity [30] (from 4.2 cP to 5500 cP, by mixing two with approximately the same thermal conductivity) resulted in decrease in the thermal conductivity of the Fe_2O_3 suspension as the viscosity of the base fluid increased. Comparative studies of SiC suspensions in water and 50/50

ethylene glycol/water mixture with controlled particle sizes, concentration, and pH showed that relative change in thermal conductivity due to the introduction of nanoparticles is ~5% higher in EG/H_2O than in H_2O [26]. This effect cannot be explained simply by the lower thermal conductivity of the EG/H_2O base fluid since the difference in enhancement values expected from EMT is less than 0.1% [15]. Therefore the "base fluid effect" observed in different nanofluid systems is most likely related to the lower value of the interfacial thermal resistance (better wettability) in the EG/H_2O nanofluids than in H_2O.

Relative viscosities of similar suspensions in EG/H_2O than in H_2O decrease with the increase of the average particle size, showing lesser viscosity increase in the EG/H_2O nanofluids, especially in suspensions of smaller nanoparticles [26]. According to the classic Einstein-Bachelor equation for hard non-interacting spheres [31], the percentage viscosity increase should be independent of the viscosity of the base fluid and only proportional to the particle volume concentration. Therefore experimentally observed change in viscosity increase in base fluids can be related to the difference in structure and thickness of immobilized fluid layers around the nanoparticles, affecting the effective volume concentration and ultimately the viscosity of the suspensions [23, 25-26].

Since both high thermal conductivity and low viscosity increases in nanofluids are important for heat transfer performance, the nanofluids prepared from more viscous base fluids will have greater potential for practical applications.

Viscosity increase in nanofludis was shown to depend not only on the type of the base fluid, but also on the pH value (in protonic fluids) that establishes zeta potential (charge at the particle's slipping plane). Particles of the same charge repel each other minimizing the particle-particle interactions that strongly affect viscosity [23, 25, 32]. It was demonstrated that the

viscosity of the alumina-based nanofluids can be decreased by 31% solely by adjusting the pH of the suspension without affecting the thermal conductivity [23]. Nanoparticles in suspensions can be well-dispersed (particles move independently) or agglomerated (ensembles of particles move together). Depending on the particle concentration and the magnitude of particle-particle interactions that are affected by pH, surfactant additives and particle size and shape, a dispersion/agglomeration equilibrium establishes in nanoparticle suspension. Extended agglomerates can provide increased thermal conductivity as described in the literature [33-34], but agglomeration and clustering of nanoparticles result in undesirable viscosity increase and/or settling of suspensions.

Introduction of other additives (salts and surfactants) may also affect the zeta potential at the particle surfaces. Non-ionic surfactants provide steric insulation of nanoparticles preventing Van-der Waals interactions, while ionic surfactants may serve as both electrostatic and steric stabilization. The thermal conductivity of surfactants is significantly lower than water and ethylene glycol. Therefore addition of such additives, while improving viscosity, typically reduces the thermal conductivity of suspension.

It should be mentioned here that all thermo-physical properties have some temperature dependence. The thermal conductivity of fluids may increase or decrease with temperature, however it was shown that relative enhancement in the thermal conductivity due to addition of nanoparticles remains constant [28, 35]. The viscosity of most fluids strongly depends on the temperature, typically decreasing with increasing temperature. It was noted in a couple of nanofluid systems that the relative increase in viscosity is reduced as temperature rises [25-26]. The fact of constant thermal conductivity increase and viscosity decrease with temperature makes nanofluids technology very promising for high-temperature application. The density and

specific heat of nanofluids change insignificantly within the practical range of current cooling applications. Stability of nanofluids could be improved with temperature increase due to increase in kinetic energy of particles, but heating also may affect the suspension stability provided by electrostatic or/and steric methods. Further studies are needed in this area.

III. Systems Engineering Approach to Nanofluids

A schematic representation of a nanofluid system (Fig. 1) illustrates the complexity of correlations between the engineering parameters and the fluid properties. Manipulation of the nanofluid system as a whole to achieve the advanced heat transfer performance requires identification of critical parameters and properties of nanofluids.

The system engineering approach used in this work is a semi-quantitative technique for ranking multi-dimensional nanofluid engineering options. It also offers an alternative way to look at the inner workings of a nanofluid system and allows for design choices addressing the heat transfer demands of a given industrial application. The available set of information on nanoparticle suspensions is arranged in a basic decision matrix (Table 1) with each engineering parameter in a column and the nanofluid properties listed in rows. Each cell in the table represents the strength of the effect of a particular parameter to the nanofluid's property with "x" indicating no dependence, "▲" – weak, "○" – medium, and "◘" for strong dependence and scored as 0.0, 0.25, 0.5 and 1.0 correspondingly. The relative importance of each nanofluid parameter can be estimated as a sum of the gained scores (Table 1). Based on that the nanofluid engineering parameters can be arranged by the decreasing importance for the heat transfer performance: particle concentration > base fluid > nanoparticle size > nanoparticle material ≈ surface charge > temperature ≈ particle shape > additives > Kapitza resistance. This is an approximate ranking of engineering parameters that assumes equal and independent weight of

each of the nanofluid properties contributing to thermal transport. The advantage of this approach to decision making in nanofluid engineering is that subjective opinions about the importance of one nanofluid parameter versus another can be made more objective.

Applications of the decision matrix (Table 1) are not limited to the design of new nanofluids, it also can be used as guidance for improving the performance of existing nanoparticles suspensions. In a given nanofluid the particle material, size, shape, concentration, and the base fluid parameters are fixed, however the cooling performance still can be improved by remaining adjustable nanofluid parameters in order of their relative importance, i.e. by adjusting the zeta potential and/or increasing the test/operation temperatures in the above case. Further studies are needed to define the weighted importance and sensitivity of each nanofluid property contributing to the heat transfer.

Summary

By analyzing the cooling efficiency criteria for single-phase fluids the properties of nanofluids that are important for heat transfer were identified. After that the nanofluid engineering parameters were reviewed in regards to their correlations to the important thermo-physical properties of nanoparticle suspensions. The system engineering approach was applied to analyze the set of nanofluid data and to identify the most influential nanofluid parameters. Based on the relative importance of engineering parameters the potential design options can be evaluated. Importantly, the criteria are not weighted to allow a quick selection process. The nanoparticle concentration, base fluid, and particle size appear to be the most influential parameters for improving the heat transfer efficiency of nanofluid.

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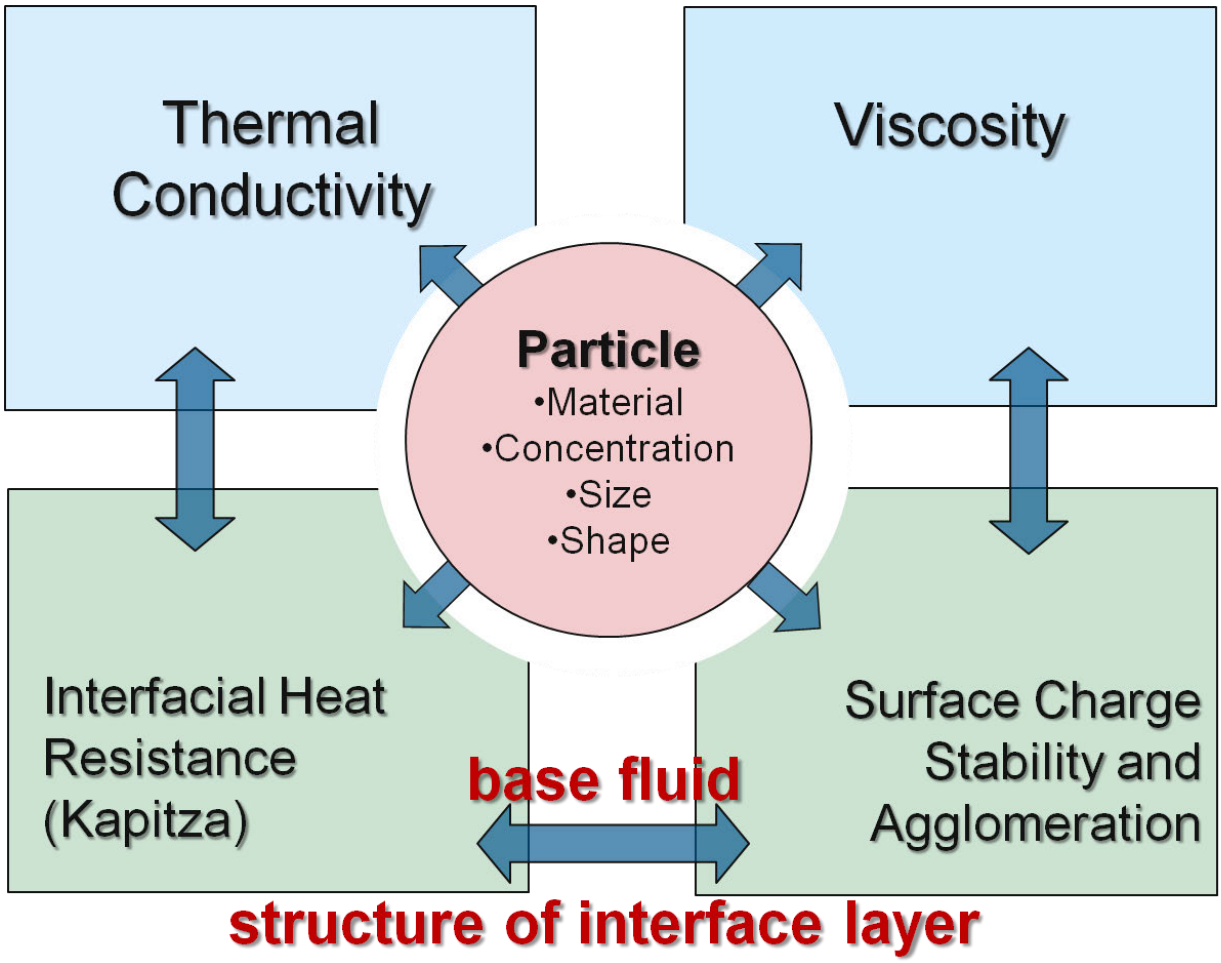

Figure 1. Schematic representation of the multivariability of a nanofluid system.

Table 1. Systems engineering approach to nanofluid design.

	ENGINEERING PARAMETERS	**Nanoparticle material**	**Nanoparticle concentration**	**Nanoparticle shape**	**Nanoparticle size**	**Base fluid**	**Zeta potential /fluid pH**	**Kapitza resistance**	**Additives**	**Temperature**
NANOFLUID PROPERTIES										
Stability	⇑	▲	▲	▲	◘↓	○	◘	X	◘	?
Density	⇑	◘	◘↑	X	X	◘	X	X	X	X
Specific Heat	⇑	◘	◘↓	X	X	◘	X	X	X	▲
Thermal Conductivity	⇑	○	◘↑	○	◘↑	▲	○	◘↓	▲	○
Viscosity	⇓	▲	◘↓	◘	◘↓	◘↑	◘	X	○	◘
Heat Transfer Coefficient	⇑	◘	◘↑*	◘	◘↑	◘	◘	◘↓	○	◘
Pumping Power Penalty	⇓	X	◘	◘	◘↑	◘	◘	X	○	◘
Relative Importance		4.0	6.25	3.75	5.0	5.25	4.0	2.0	2.75	3.75

Symbols:

◘ - strong dependence; ○ - medium dependence; ▲ - weak dependence; X - no dependence; ? – unknown or varies from system to system; ⇑ - larger the better; ⇓ - smaller the better; ↑ - increase with increase in parameter; ↓ - decrease with increase in parameter; *- within the linear property increase.