



Nanofluids: Stability, phase diagram, rheology and applications

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ABSTRACT

There is no doubt about the potential technological significance of nanofluids. The promising application areas have been identified as effective heat transfer fluids, contrast agents in magnetic resonance imaging, magnetohyperthermia treatment, precursors to high performance nanocomposites and ordered nanostructures. However, commercial applications are rare, in part due to the limited understanding of the nanofluid fundamentals such as colloid stability, phase diagrams and rheology. This paper intends to provide a brief overview of the scientific disciplines that are important to nanofluids, and the interconnection among different disciplines in order to gain a perspective on the future development of this intriguing area.

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1. Introduction

A nanofluid refers to a fluid that contains particles with dimensions less than 100 nm. The base fluid, or dispersing medium, can be aqueous or non-aqueous in nature. Typical nanometer-sized particles are metals, oxides, carbides, nitrides or carbon nanotubes. Their shapes may be spheres, discs or rods. The study of nanofluids is an important branch of nanotechnology, since it has potential applications in transportation and electronics cooling (Buongiorno et al., 2009; Eastman, Choi, Li, Yu, & Thompson, 2001; Godson, Raja, Mohan Lal, & Wongwises, 2010; Shalkevich, Shalkevich, & Bürgi, 2010; Wang & Mujumdar, 2007; Wu et al., 2010), biomedicine (Ai et al., 2005; Grainger & Castner, 2008; Ma & Liu, 2007; Nasongkla et al., 2006; Xu, Li, Li, Feng, & Gao, 2009), oil drilling (Ju, Fan, & Ma, 2006), nano-structure fabrication (Ai et al., 2005; Choi & Alivisatos, 2010; DeVries et al., 2007; Kane, Inan, & Saraf, 2010; Nasongkla et al., 2006; Rao et al., 2009; Tam et al., 2010) and nanocomposites (Rao & Chen, 2008; Guo, Han, Li, & Zhao, 2010; Lu, Liu, & Lee, 2005). Its fundamental science spans colloidal science, surface chemistry, fluid mechanics and materials science. It is this paper's intent to provide a brief overview of the scientific disciplines that are important to nanofluids, as the past few years have seen significant advances in both related disciplines and applications. Furthermore, this paper aims to illustrate the interconnection among different disciplines and provide perspective to accelerate the future development of this intriguing area of study.

2. Colloidal stability

When nanoparticles are dispersed in a base fluid, the state of the nanoparticles is determined by the interaction between the nanoparticles and between the nanoparticle and base fluid. The colloid stability in water is best studied using DLVO theory (Deryaguin & Landac, 1941; Overbeek, 1952; Verwey & Overbeek, 1948). Although a more recently proposed extended DLVO theory describes better the stability of nanofluids (Boström, Deniz, Franks, & Ninham, 2006), a good theoretical framework and analytical tools to support the development of stable nanofluids are still lacking.

Basic DLVO theory mainly concerns the forces between two particles in water: the attraction force, such as van der Waals force, and the primary repulsion force, known as the electrical double layer that results from charge interactions. The van der Waals interaction potential profile (potential vs. distance away from the particle surface) is as follows (Hamaker, 1937):

$$V_A = -\frac{A}{12} \left[\frac{1}{x(x+2)} + \frac{1}{(x+1)^2} + 2 \ln \frac{x(x+2)}{(x+1)^2} \right], \quad \text{where } x = \frac{h}{2a}. \quad (1)$$

In Eq. (1), h is the distance between the surfaces of two particles, a is the radius of the particle, and A is the Hamaker constant, which describes the attraction strength of the two particles in a medium. The “–” sign indicates the attraction nature of the potential. The Hamaker constant scales with polarizability and density and has a unit of erg. The attraction of the medium also reduces the overall attraction force between two particles. For instance, the Hamaker constant of cyclohexane is higher than that of water. Therefore, the

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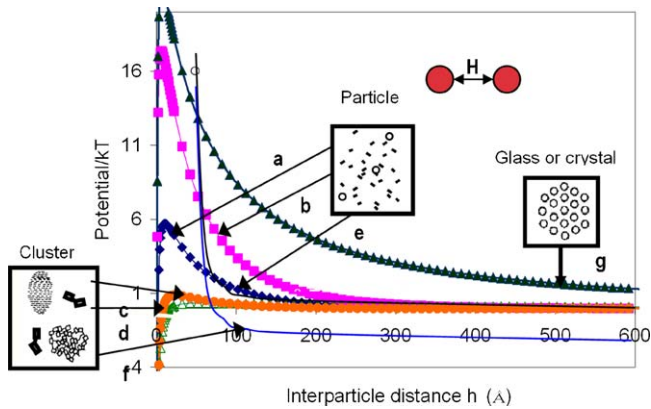


Fig. 1. Potential curves and the state of nanoparticles in a nanofluid. The legends of the curves are: (a) open diamond; (b) open square; (c) solid circle; (d) solid triangle; (e) open ellipsoid; (f) open cross; (g) open triangle. The radius of the nanoparticle $a = 25 \text{ \AA}$ and a Hamaker constant $A = 5 \times 10^{-13} \text{ erg}$.

attraction force of two silica particles in cyclohexane is smaller than that in water (Bergström, 1997).

When the particles dispersed in the medium are charged, electrical double layer forms. The zone of ion diffusion is characterized by the Debye length. This is an important mechanism to prevent the aggregation of particles by van der Waals attraction. The charge interaction between two identical particles can be described as follows for a constant charge:

$$V_R^\psi = \frac{\varepsilon a \psi_0^2}{2} \ln(1 + \exp(-\kappa h)), \quad (2)$$

where ε is the dielectric constant of the medium, ψ_0 is the particle surface charge potential, and κ is the reciprocal of the Debye length, defined as

$$\kappa = \left(\frac{8\pi e^2 n z^2}{\varepsilon k T} \right)^{1/2} \quad (3)$$

where e is elemental electron charge, n is the number of ions per cm^3 , and z is the valence of the ion.

The stability of a colloid fluid is determined by the summation of the attraction and repulsion forces. If the energy barrier is insufficient to prevent the system from achieving a lower energy state, the attraction of the van der Waals forces will prevail and particles will aggregate and settle out of the base fluid (Cosgrove, 2005; Hiemenz & Rajagopalan, 1997). Eqs. (2) and (3) highlight the importance of particle length scale on the stability of a colloidal system. The stability of even a seemingly simple emulsion such as paint, with micron-sized particles dispersed in water, remains a challenge in its application. Many commercial polymeric dispersants help stabilize pigments such as Dow's TamolTM, AcumerTM, and OrotanTM, all acid based copolymers. Such challenge is multiplied for nanofluids, as the individual particles are smaller than typical latexes by two orders of magnitude. The stabilization of nanoparticles becomes more challenging when nanoparticles are dispersed in non-aqueous media such as ethylene glycol and the dielectric constant of a non-aqueous medium is much lower than that of water (typically $< 1/10$). Several potential profiles are shown in Fig. 1, where the curves are calculated from parameters defined in Table 1.

The stability of an aqueous nanofluid is demonstrated in Fig. 1 Curves a, b, and c. When a surface potential is 45 mV (Curve a), the nanoparticles assume a single particle state. The higher the particle charge potential, the more stable is the nanofluid, as shown in Curve b. Generally, nanofluids are kinetically stable systems because the particles can move over a small distance undergoing

Table 1

Parameters used to calculate the potential curves in Fig. 1.

Potential curve legend	ε	ψ_0 (mV)	κ ($1/\text{\AA}$)	D (\AA)
a	80	45	0.008	0
b	80	70	0.008	0
c	80	25	0.008	0
d	5	70	0	0
e	5	70	0	30
f	5	70	0	30
g	80	45	0.0008	0

D is the thickness of an adsorbed shell on the particle.

Brownian motion where van der Waals attraction prevails and thus aggregation occurs (Qi, Wu, Zhang, Zeng, & Zhi, 2010). As shown in Curve c, if the surface charge is not high enough, there is essentially no barrier and particles will aggregate to form different secondary structures. Once the secondary structures start to grow, the energy balance changes due to the change in particle size and the system can be stable in an intermediate state where the particles remain suspended at some small-sized secondary structure (Zhang, González-Mozuelos, & Olvera de la Cruz, 2010).

In a non-aqueous medium, the repulsion force is greatly reduced resulting in more aggregated nanoparticles, as shown in Curve d. A general strategy for stabilizing non-aqueous nanofluids is to surface coat the nanoparticles, typically with a layer that is polymeric in nature. There are two stabilizing effects that result from this strategy; the first effect is due to the change in particle size, while the second effect is the decrease in attraction force. These effects are illustrated in Curve e, where a 3 nm thick coating layer serves as an energy wall that effectively "screens" the attraction force and stabilizes the nanoparticles (Adireddy, Lin, Cao, Zhou, & Caruntu, 2010; Chou, Oyler, Motl, & Schaak, 2009; Guo et al., 2010; Heath, Shiang, & Alivisatos, 1994; Olshavsky & Allcock, 1997; O'vařri, Berkoř, Balařz, Majzik, & Kiss, 2010; Pastoriza-Santos & Liz-Marzán, 2009; Shchukin & Sukhorukov, 2004). The coating material can also introduce charges that increase electrostatic repulsion, further stabilizing the nanoparticles (Bucher et al., 2002; Yoonessi, Seikel, & Pender, 2009). In situ stabilization, where the surface stabilization groups are introduced during nanoparticle synthesis, is also commonly practiced (Adireddy et al., 2010; Bucher et al., 2002; Chou et al., 2009; Heath et al., 1994; Olshavsky & Allcock, 1997; Pastoriza-Santos & Liz-Marzán, 2009; Shchukin & Sukhorukov, 2004; Yoonessi et al., 2009). The surface stabilization groups not only create a protective shell, but also direct and control the particle growth.

A system undergoing depletion flocculation is shown in Curve f of Fig. 1. Flocculation can happen when excessive surfactant or polymers are present in a nanofluid. In this case, forces exist in addition to van der Waals attraction and electrostatic repulsion. One of these forces is the attraction between particles due to volume restriction. Entropy arguments suggest that surfactant molecules are less likely to be present between two particles than in the bulk solution. Another force is the osmosis pressure caused by the incompressibility of the surfactant chain (Asakura & Oosawa, 1954; Fan & Tuinier, 2010; Fresnais, Lavelle, & Berret, 2009; Heller, 1980; Lyklema, 1968; Schowalter & Eidsath, 2001; Vrij, 1976; Wang, Sarkar, & Nicholson, 1997).

Thus far, colloidal stability has been discussed, and related potential curves are shown in Fig. 1, Curves a–f. However, in some cases repulsion dominates attraction and even long-range repulsion may be present in the nanofluid. In such systems, as shown in Fig. 1, Curve g, the Debye length is sufficiently larger that the nanoparticles have arranged themselves in an orderly fashion to maximize their available states. The nanofluid can lose flowability in such cases, creating difficulty in situations where low

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