Structure – Property Relationships for Nanofluids

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Abstract Nanofluids refer to dilute liquid suspensions of nanoparticles in commonly used heat transfer liquids. They triggered much excitement since mid 1990s mainly owing to the claims of anomalous enhancement of thermal conductivity even at very low nanoparticle concentrations. There have been numerous attempts to interpret the mechanism(s) that drive the displayed enhancement. A long debate within the research community supported by experimental and theoretical evidence has highlighted the nanoparticle structuring as the dominant underlying mechanism. On the other hand the viscosity increase as a result of nanoparticle structuring raises concerns about their suitability for certain applications. This paper mainly discusses the structure – property relationship for nanofluids in microscopically static conditions.

Keywords: Nanofluids, Particle Structuring, Rheology, Thermal Conductivity, Structure - Property Relationship

1. Introduction

Nanofluids are made of a base liquid and a dissimilar material in the form of nanoparticles. The terminology 'nanofluids' was first put forward by Dr Stephen Choi (Choi et al. 1995) and the initial stage of research on the subject was mainly conducted at the Argonne National Laboratory USA with a focus on thermal conductivity under macroscopically static conditions. The topic gained worldwide attention from late 1990s and became very popular from around 2002 as indicated by the exponential growth in the number of publications. The popularity of the topic of nanofluids is associated with some early experimental observations of enhanced properties and behaviour in heat transfer (Choi et al. 2001), mass transfer (Krishnamurthy et al. 2006, Olle et al. 2006), wetting and spreading (Wasan and Nikolov 2003), and antimicrobial activities (Zhang et al. 2007). This paper deals with the fundamental transport properties of nanofluids, in particular the thermal conductivity and viscosity. The focus is on understanding how the nanoparticle structuring affect the two transport properties and whether the two properties can be related

via the structural information. The paper is organised in the following manner; Section 2 reviews briefly the work on thermal conductivity and viscosity of nanofluids where the effect of nanoparticle structuring on the transport properties is also discussed. An attempt is made in Section 3 to relate the thermal conductivity and viscosity through nanoparticle structuring. Finally, a summary is given in Section 4.

2. Thermal conductivity and viscosity of nanofluids

2.1 Thermal conductivity of nanofluids

Investigations on the thermal conduction of nanofluids under macroscopically static conditions dominated the early stage of nanofluids research. There were few reviews published over the last decade, for example by Keblinski et al. (2005), Das et al. (2006), Kabelac and Kuhnke (2006), Wang and Mujumdar (2007), Ding et al. (2007a) and Yu et al. (2008). Representative data on thermal conductivity of nanofluids at room temperature are plotted on figure 1. These were extracted from Lee et al. (1999), Eastman et al. (2001), Choi et al. (2001), Xie et al.

(2002a & 2002b), Biercuk et al. (2002), Das et al. (2003a), Patel et al. (2003), Kumar et al. (2004), Assael et al. (2004), Zhang X. et al. (2007), Wen and Ding (2004a, 2004b, 2005a, 2005b, 2006), Ding et al. (2006) and He et al. (2007).
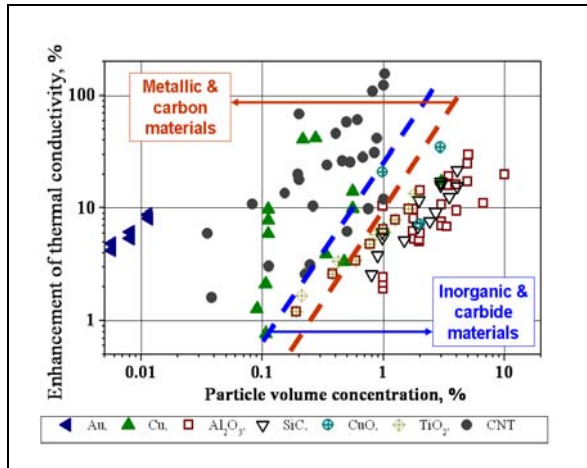

Figure 1: Measured thermal conductivities of nanofluids

On abscissa of figure 1 are the particle volume fractions ($\phi\%$) and on ordinate are the percentage enhancement ($k_{nf}*100/k_l$), where k_{nf} and k_l are thermal conductivities of nanofluids and base liquids respectively.

Obviously the experimental data shows a significant scattering. Nevertheless, there is clear evidence to show the presence of nanoparticles have enhanced the thermal conductivity of liquids in many cases. The degree of enhancement depends on the material type and volume fraction; higher volume fraction gives greater enhancement.

A number of mechanisms have been proposed to interpret the thermal conductivity data. These include Brownian motion of nanoparticles (Patel et al. 2003, Kumar et al. 2004, Koo and Kleinstreuer 2004, Prasher et al. 2006a), interfacial ordering of liquid molecules at nanoparticle surfaces (Keblinski et al. 2002, Yu and Choi 2003, Wang et al. 2003), ballistic transport of energy carriers within individual nanoparticles (Keblinski et al. 2002), as well as nanoparticle structuring / networking (Keblinski et al. 2002, Wang et al. 2003, Nan et al. 2003, Prasher et al. 2006b & 2006c). The ballistic transport of energy

carriers occurs when the mean free path of the energy carriers is larger than the characteristic length of nanoparticles. Such a mechanism has been excluded as a possible mechanism for the enhanced thermal conductivity because the thermal conductivity of nanoparticles decreases with decreasing particle size when the size becomes comparable to the mean free path of the energy carriers (Chen 1996).

Brownian motion of nanoparticles could contribute to the thermal conduction enhancement through two ways, direct contribution due to motion of nanoparticles that transport heat, and indirect contribution due to micro-convection of fluid surrounding individual nanoparticles. The direct contribution of Brownian motion has been shown theoretically to be negligible as the time scale of the Brownian motion is about 2 orders of magnitude larger than that of the thermal diffusion in the base liquid (Keblinski et al. 2002). The indirect contribution has also been shown to play a minute role through the same reasoning for the direct contribution and also through molecular modelling (Evans et al. 2006). Furthermore, nanoparticles are often in the form of agglomerates and / or aggregates, so the Brownian motion is expected to play a much smaller role.

At a solid-liquid interface, liquid molecules could be significantly more ordered than that in the bulk liquid. By analogy to the thermal behaviour of crystalline solids, the ordered structure could give thermal conductivity enhancement (Keblinski et al. 2002). Such a suggestion inspired a number of researchers to develop theoretical models to explain the experimentally observed thermal conduction enhancement. However, it is now clear that liquid-nanoparticle interface is one of the main factors that lead to a decrease (rather than increase) in the effective thermal conductivity due to the so-called Kapitza interfacial resistance (Shenogin et al. 2004a & 2004b, Nan et al. 2003, Gao et al. 2007), and the effect of interfacial resistance on the overall effective thermal conductivity depends on particle size (Keblinski et al. 2005, Prasher et al. 2005, Putnam et al. 2006, Gao et al. 2007). When particle size is relatively small in

comparison with the characteristic length scale for the interfacial resistance, nanoparticles act as insulators. This can lead to deterioration of the thermal conduction of nanofluids (Putnam et al. 2006, Zhang X. et al. 2007).

Recent studies have suggested that nanoparticle structuring / aggregation be a dominant mechanism for the experimentally observed thermal conductivity enhancement of nanofluids (Wang et al. 2003, Nan et al. 2003, Prasher et al. 2006b & 2006c, Hong et al. 2006, Chen et al. 2007a, Keblinski 2008, Chen et al. 2009 & 2009b). Using such a mechanism, well-dispersed nanoparticles in a fluid matrix gives the lowest thermal conductivity, whereas interconnected nanoparticles in the liquid enhances the thermal conduction. This can be understood from the viewpoint of circuit analyses; the well-dispersed situation is closer to conductors connected in a series mode, while the interconnected case is closer to those in a parallel mode (Keblinski et al. 2008). It should however be noted that although nanoparticle structuring is able to explain the experimentally observed thermal conduction enhancement, it does not necessarily mean nanoparticle structuring is the only dominant mechanism.

2.2. Rheological behaviour of nanofluids

Only a small number of studies have been found in the literature on the rheological behaviour of nanofluids and there are inconsistencies among them. Main findings of these studies are discussed below.

Are nanofluids Newtonian or non-Newtonian? There have been debates on shear-dependent behaviour of nanofluids. The Newtonian behaviour has been observed by Wang et al (1999) and Das et al. (2003b) for water-Al_2O_3 nanofluids, Wang et al (1999) for EG-Al_2O_3 nanofluids, Xuan and Li (2003) for Cu-water nanofluids and Prasher et al. (2006d) for Al_2O_3-PG (Propylene glycol) nanofluids, whereas the non-Newtonian behaviour of nanofluids has been reported by Kwak and Kim (2005), Ding et al (2006), Ding et al. (2007b), He et al. (2007), and Chen et al. (2007b, 2008). Detailed rheological analyses

show that nanofluids can exhibit either or both Newtonian and non-Newtonian behaviour depending on particle size and shape, particle concentration, base liquid viscosity, and solution chemistry etc. The shear-dependence of viscosity of very dilute nanofluids is negligible, while nanofluids with relatively high particle concentrations are more likely to exhibit shear thinning behaviour. Nanofluids made of low viscosity fluids (e.g. water) are more likely to exhibit the non-Newtonian behaviour than those made of highly viscous base fluids (e.g. ethylene glycol or propylene glycol). Particle shape plays a strong effect on the shear dependent behaviour of nanofluids. Nanofluids containing rod-like nanoparticles, particularly rods with large aspect rations, are most likely to show strong shear thinning behaviour. For highly concentrated suspensions containing non-spherical particles, shear thickening may occur at high shear rates; see for example Egres and Wagner (2005). This is beyond the normal concentration range for nanofluids for heat transfer applications as the penalty due to viscosity increase outweighs the benefits of heat transfer performance increase.

How does temperature affect the viscosity of nanofluids? The temperature dependence of viscosity of nanofluids has been investigated experimentally by Praveen et al. (2007), Chen and Ding (2007a), Nguyen (2008) and Chen et al. (2009a & 2009b). These studies show that temperature can have a very strong effect on the shear viscosity of nanofluids. At low shear rates, the viscosity of nanofluids increases with increasing temperature, exhibiting a stronger shear-thinning behaviour (Chen et al. 2007b, 2009b). In high shear rate region where the shear viscosity approaches a constant value (called high shear viscosity), the viscosity scales with temperature in a similar fashion to that of the base liquid. The experimental results also show that both the high shear viscosity of nanofluids and the base liquids follow well the classical Vogel-Tammann-Fulcher (VTF) equation as pointed out by Chen et al. (2009b). Interestingly, however, the relative increment of the high shear viscosity at a given particle concentration is

almost independent of temperature (Chen et al. 2007b & 2009b).

How do particle size and shape affect the viscosity of nanofluids? Very few studies have been reported in literature on the effects of particle size and shape on the rheological behaviour of nanofluids. He et al. (2007) showed experimentally that an increase in particle size led to an increase in the shear viscosity, whereas Chen et al. (2009b) demonstrated that nanofluids containing rod-like particles had a much stronger shear thinning in comparison with those containing spherical nanoparticles.
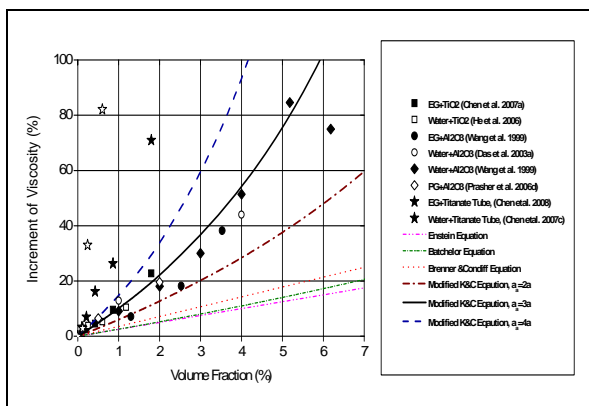

Figure 2 High shear viscosity of nanofluids as a function of nanoparticle concentration

Can the high shear viscosity of nanofluids be predicted? For shear thinning nanofluids, the shear viscosity approaches a constant at high shear rates as mentioned above. Such a constant is termed the high shear viscosity. This parameter is very relevant to convective heat transfer applications where heat transfer fluids are often in vigorous motion and subject to very high shear. Figure 2 summarises the room temperature data of high shear viscosity reported in the literature (Wang et al. 1999, Das et al. 2003a, Prasher et al. 2006d, Chen et al. 2007a & 2007b, He et al. 2007, Chen et al. 2008) together with predictions by various classical models in their original forms including Einstein equation (Einstein 1906 and 1911) for dilute non-interacting suspensions of spherical particles, Batchelor equation (Batchelor 1977) for semi-dilute suspensions of spherical particles and Brenner-Condiff

equation (B-C equation) of Brenner and Condiff (1974) for dilute suspensions of rod-like particles. In spite of the data scattering, one can see that the presence of nanoparticles in fluids increases the high shear viscosity and the extent of increment depends on nanoparticle shape and volume fraction. Given a volume fraction of nanoparticles, the high shear viscosity of nanofluids containing rod-like particles is much higher than those containing spherical nanoparticles. Given particle shape, the high shear viscosity of nanofluids increases with increasing nanoparticle volume fraction in a nonlinear manner and the original form of the classical models mentioned above fail to provide an adequate prediction by a large margin,. Theoretical analyses using the classical colloidal theories have indicated that nanoparticle structuring is able to explain the experimentally observed high-shear viscosity increase; see for example Chen et al. (2007a, 2007b, 2009a, 2009b).

3. Relationship between thermal conductivity and rheological behaviour of nanofluids

As discussed above, current experimental and theoretical evidence suggests nanoparticle structuring could be a likely dominant mechanism for the experimentally observed thermal conduction enhancement for nanofluids. Experimental and theoretical analyses further suggest that nanoparticle structuring is responsible for the viscosity increase. Hence there could be a nexus between the thermal conductivity and rheological behaviour of nanofluids. This is explained using the nanofluids containing spherical particles as follows.

From the rheological behaviour, the effective size of structured (aggregated) nanoparticles and the effective volume fraction of nanofluids can be estimated; see Chen et al. (2007b, 2009b). Such information can then be used to modify the conventional Hamilton-Crosser (H-C) model (Hamilton Crosser 1962), which would give the effective thermal conductivity. Note that the thermal conductivity of aggregates needed to feed the H-C model is obtained from Bruggeman model (Bruggeman 1935). Figure 3 compares

the calculated thermal conductivity based on the above procedure and those obtained from experiments. A reasonably good agreement has been found.
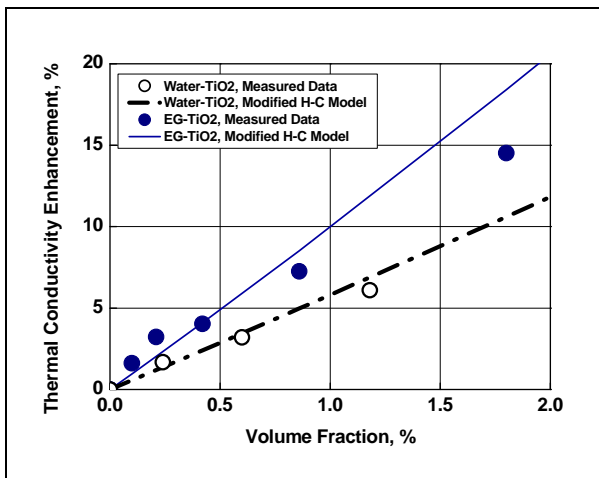

Figure 3 Comparison between calculated thermal conductivity enhancement based on rheological data and experimental results for nanofluids containing spherical titania particles

4. Concluding remarks

This paper shows how nanoparticle structuring affects the transport properties of nanofluids and the possibility of relating the thermal conductivity and viscosity of nanofluids through nanoparticle structuring. Such a methodology could enable design of nanofluids with optimal performance for heat transfer and other applications. To make it a reality, the following challenges need to be addressed:

(a) Most applications involve flows of nanofluids. It is unclear how nanoparticles structure under dynamic flow conditions.

(b) There is a lack of method for tuning the structures of nanoparticles under dynamic conditions

(c) In heat transfer conditions, the temperature fields are non-uniform. It is unclear how temperature affects nanoparticle structuring.

(d) Although the order of magnitude of interfacial resistance is known for a small number of materials, this is not sufficient to give predictive capabilities.

These challenges are mostly engineering in nature but require significant effort to resolve. Some of them might never get resolved as benefits may be outweighed by the costs.

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