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Rheological behaviour of nanofluids

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Abstract. This work aims at a more fundamental understanding of the rheological behaviour of nanofluids and the interpretation of the discrepancy in the recent literature. Both experiments and theoretical analyses are carried out with the experimental work on ethylene glycol (EG)-based nanofluids containing 0.5-8.0 wt% spherical TiO₂ nanoparticles at 20-60 °C and the theoretical analyses on the high shear viscosity, shear thinning behaviour and temperature dependence. The experimental results show that the EG-based nanofluids are Newtonian under the conditions of this work with the shear viscosity as a strong function of temperature and particle concentration. The relative viscosity of the nanofluids is, however, independent of temperature. The theoretical analyses show that the high shear viscosity of nanofluids can be predicted by the Krieger–Dougherty equation if the effective nanoparticle concentration is used. For spherical nanoparticles, an aggregate size of approximately 3 times the primary nanoparticle size gives the best prediction of experimental data of both this work and those from the literature. The shear thinning behaviour of nanofluids depends on the effective particle concentration, the range of shear rate and viscosity of the base liquid. Such non-Newtonian behaviour can be characterized by a characteristic shear rate, which decreases with increasing volume fraction, increasing base liquid viscosity, or increasing aggregate size. These findings explain the reported controversy of the rheological behaviour of nanofluids in the literature. At temperatures not very far from the ambient temperature, the relative high shear viscosity is independent of temperature due to negligible Brownian diffusion in comparison to convection in high shear flows, in agreement with the experimental results. However, the characteristic shear rate can have strong temperature dependence, thus affecting the shear thinning behaviour. The theoretical analyses also lead to a classification of nanofluids into dilute, semi-dilute, semi-concentrated and concentrated nanofluids depending on particle concentration and particle structuring.

Symbol

- *A* constant coefficient
- *a* radius of particle
- $a_{\rm a}$ effective radius of aggregates
- *B* constant coefficient
- *C* constant coefficient
- *k*_B Boltzmann conductivity
- $k_{\rm H}$ Huggins's coefficient
- *m* power law index
- N test number
- *n* power law index
- *Pe* Péclet number
- *T* absolute temperature
- t temperature

Greek

	00
φ_{a} effective volume concentration of aggregation	62
<i>w</i> particle concentration by weight	
$\dot{\gamma}$ shear rate	
σ shear stress	
η_0 viscosity of base liquid	
η viscosity of suspension	
η_i viscosity increase	
$\eta_{\rm r}$ relative viscosity	

 $[\eta]$ intrinsic viscosity

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1. Introduction

Nanofluids are dilute liquid suspensions of nanoparticles with at least one of their critical dimensions smaller than about 100 nm (Choi 1995). Much attention has been paid in the past decade to this new type of composite materials because of their enhanced properties and behaviour associated with heat transfer, mass transfer, wetting and spreading and antibacterial activity. These enhanced properties and behaviour imply an enormous potential of nanofluids in device miniaturization and process intensification which could have an impact on many industrial sectors including chemical and process, transportation, electronics, medical and energy and environment.

Most published studies on nanofluids deal with the heat transfer behaviour including thermal conduction (Choi 1995; Masuda *et al* 1993; Wen and Ding 2004a), phase change (boiling) heat transfer (Das *et al* 2003a, 2003b; Wen and Ding 2005a) and convective heat transfer (Ding *et al* 2006; Pak and Cho 1998; Wen and Ding 2004b, 2005b; Yang *et al* 2005), which have been reviewed recently by Keblinski *et al* (2005) and Das *et al* (2006). Very few studies, however, have been reported on the rheological behaviour of nanofluids (Kwak and Kim 2005; Prasher *et al* 2006; Tseng and Lin 2003) although there is a large body of rheology literature on suspensions and/or colloids. Nanofluids can be regarded as functionalized colloids. Key requirements for nanofluids include low solids loading but with high thermal properties, favourable rheologocial properties and flow behaviour, and great stability over a wide range of temperature to meet the industrial needs. Clearly, there is a gap in the current rheological literature for this type of fluids. This forms the primary motivation of this work.

Recent work has shown evidence of a relationship between the thermal and rheological behaviour of nanofluids (Ding *et al* 2006; Lee *et al* 2006; Yang *et al* 2006). The questions are: how are the two related? Does the relationship follow the convectional fractal structure based theory? Can the relationship explain the experimentally observed thermal behaviour of nanofluids? Are there new physics involved? Given the recent surge in nanofluids research, there is an urgent need to answer these questions. This forms the second motivation of carrying out this work.

This work aims at understanding the rheological behaviour of nanofluids through both experimental work and theoretical analyses. The focus will be on nanofluids containing spherical particles. The experimental work uses the ethylene glycol (EG)-based titania (TiO₂) nanofluids, whereas the theoretical analyses is based on our current understanding of the rheology of colloid suspensions. The reasons for the use of EG-based TiO₂ nanofluids are: (i) TiO₂ is generally regarded as a safe material for humans and animals although it is recognized that this may change in the future with more fundamental research on nano-toxicology; (ii) TiO₂ nanoparticles are produced on large industrial scales so are easy to obtain; (iii) metal oxides such TiO₂ nanoparticles are chemically more stable than their metallic counterparts although it is recognized that the photocatalytic effect of TiO₂ may impose restrictions in selecting the base liquid; (iv) EG can be used over a fairly wide range of temperature. Two additional reasons for the work are as follows.

1. Despite a couple of studies on the thermal properties of EG-based nanofluids (Wang *et al* 1999), little is found in the literature on the rheological behaviour of EG-based TiO₂ nanofluids. Although Chadwick *et al* (2002) studied the rheological behaviour of suspensions of uncoated anatase TiO₂ in EG and suggested that TiO₂ is in the form of aggregates with a size of ~600 nm, the primary particle size was not given. Also, the work



Figure 1. SEM micrograph of TiO₂ nanoparticles as received.

by Chadwick *et al* (2002) did not examine the effect of temperature, which is fundamentally important to nanofluids for heat transfer applications (Abdulagatov *et al* 2006; Prasher *et al* 2006).

2. There are discrepancies in the reported studies of nanofluids. Some concluded that nanofluids were Newtonian (Das *et al* 2003; Prasher *et al* 2006; Wang *et al* 1999; Xuan and Li 2000); others found non-Newtonian behaviour of nanofluids (Ding *et al* 2006; He *et al* 2007; Kwak and Kim 2005; Studart *et al* 2006; Tseng and Lin 2003). There is an urgent need to address this issue.

The paper is structured in the following manner. Section 2 describes briefly the experimental methods. The results are presented and discussed in section 3. Analyses of the rheological behaviour of nanofluids will be performed in section 4, where interpretation of the discrepancies reported in the literature will also be made. Review of the recent literature is included where appropriate.

2. Experimental

EG-based TiO₂ nanofluids were formulated with dry TiO₂ nanoparticles (P25, Degussa, Germany) and pure EG (Alfa Aesar, USA) by using the so-called two-step method (Wen and Ding 2005c; Zhu *et al* 2004). Figure 1 shows an SEM image of dry TiO₂ nanoparticles. It can be seen that the primary nanoparticles are spherical with approximately 25 nm diameter. However, they are in the form of large agglomerates. Nanofluids with 0.5, 1.0, 2.0, 4.0 and 8.0% by weight were formulated, which corresponded respectively to 0.10, 0.21, 0.42, 0.86 and 1.80% by volume. No dispersant/surfactant was used in the formulation to avoid complication in interpreting the experimental results. The effectiveness of such an approach can be seen from figure 2, which plots the average particle size (diameter) against the processing time for 2.0 wt% nanofluids, where particle size was measured by a Malvern Nanosizer



Figure 2. Average particle size as a function of ultrasonification processing time.

(Malvern Instrument, UK). It can be seen that the average size decreases rapidly in the first 20 h after which the curve levels off with a value of \sim 140 nm. The processing time was therefore set for 20 h for making all the nanofluids. The average particle size for nanofluids containing 1.0, 2.0, 4.0 and 8.0% by weight were measured to be in the range 140–150 nm, whereas that containing 0.5 wt% was \sim 120 nm. It should be noted that the Malvern Nanosizer measures hydrodynamic properties based on the Stokes–Einstein equation (Goodwin 2003), which is expected to be slightly larger than the actual size.

Nanofluids produced were found to be very stable and there was no visually observable sedimentation after more than two months. This was also confirmed by particle size measurements over the period of two months. As mentioned above, the measured average particle size in the formulated nanofluids is much larger than the size of primary particles. This indicates that the 2-step method was not able to break the agglomerates into primary particles. This is confirmed by SEM analyses, which show that the size of agglomerates after ~ 20 h sonication is 50–150 nm with most of them in the range of 70–100 nm; see figure 3.

The formulated nanofluids were subjected to rheological analyses over a temperature range of 20–60 °C using a Bolin CVO rheometer (Malvern Instruments, UK). The measurements were based on the controlled shear stress model with the stress ranging from 0.05 to 5 Pa. Calibrations were conducted against standard solutions on a weekly basis over the duration of this work to ensure accurate measurements of the viscosity of nanofluids. The maximum uncertainty was found to be $\sim 1.7\%$.

3. Experimental results

Initial experiments were carried out on the base liquid of pure EG. Figure 4 shows the shear rate $(\dot{\gamma})$ as a function of shear stress (σ) (figure 4(a)) and viscosity (η) as a function of shear rate (figure 4(b)) at 20, 30 and 40 °C. It can be seen that the shear rate depends linearly on the shear stress and the viscosity is a constant, indicating the Newtonian behaviour of EG under



Figure 3. SEM micrograph of a nanofluid (1.0 wt.%/0.21 vol.%).



Figure 4. Rheological behaviour of pure EG at 20 °C, 30 °C and 40 °C.

the conditions of this work. The shear viscosity at $20 \,^{\circ}$ C is approximately twice that at $40 \,^{\circ}$ C (figure 4(b)), indicating a strong temperature effect on the viscosity of EG. This is demonstrated more clearly in figure 5 where the shear viscosity of EG is plotted against temperature. Also included in figure 5, are three data points of Sun and Teja (2003). Regression of the data gives the following expression:

$$\ln \eta_0 = A + B \times 1000/(T + C), \tag{1}$$

where η_0 is the shear viscosity of EG (mPa s), *T* is the absolute temperature (K), and *A*, *B* and *C* are constants given respectively by -3.2114, 0.86973 and -154.57. Equation (1) takes a similar format to that widely used for liquid viscosity (Bird *et al* 2002). It can be seen that the measured data agree very well with those reported by Sun and Teja (2003) and equation (1) fits very well to the experimental data under the experimental conditions of this work. The quality of fitting

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Figure 5. Viscosity of pure EG as a function of temperature.

Table 1. Discrepancies between measurements and empirical models for pure EG.

Comparison	А	В	С	MaxD ^a	$\operatorname{MinD}^{\mathrm{b}}$	AAD ^c	GAD ^d
Sun's/equation (1)				+0.81%	+0.03%	+0.41%	0.52%
Current/equation (1)	-3.2114	0.86973	-154.5686	+0.62%	-1.44%	-0.40%	0.78%

^aMaximum discrepancies.

^bMinimum discrepancies.

^cArithmetic averaged discrepancies.

^dGeometric averaged discrepancies.

is shown in table 1; one can see that the maximum discrepancy between equation (1) and the experimental data is $\sim 1.6\%$.

Having established confidence with the base liquid, the rheological behaviour of the formulated nanofluids were measured over a temperature range of 20-60 °C (293.15-333.15 K) with an interval of 5 K. Figure 6(a) shows the shear rate as a function of shear stress for 0.5, 2.0 and 8.0 wt% nanofluids at 30 °C. The results for other concentrations are similar. The linear relationship between shear rate and shear stress indicates the Newtonian behaviour of the EG-based TiO₂ nanofluids at a shear rate higher than 0.05 s^{-1} for all concentrations and temperatures investigated. This is more clearly shown in figure 6(b), where the viscosity is plotted against the shear rate for the 2.0 wt% nanofluids at 20, 30 and 40 °C. The Newtonian behaviour has also been observed by Wang *et al* (1999) and Das *et al* (2003) for water–Al₂O₃ nanofluids, Wang *et al* (1999) for EG–Al₂O₃ nanofluids, Xuan and Li (2000) for water–Cu nanofluids and Prasher *et al* (2006) for propylene glycol (PG)–Al₂O₃ nanofluids. However, these studies were carried out at relatively high shear rates at which the non-Newtonian behaviour is not obvious. This work covers a wide range of shear rate ($0.05-10000 \text{ s}^{-1}$) and the observed Newtonian behaviour is somewhat surprising as our previous work on water–TiO₂ nanofluids showed a strong non-Newtonian behaviour at shear



Figure 6. Rheological behaviour of nanofluids.



Figure 7. Viscosities of nanofluids at different temperatures.

rates lower than $\sim 100 \text{ s}^{-1}$, where TiO₂ nanoparticles were the same as those used in the current work (He *et al* 2007). These controversies suggest that the properties of the base liquid and nanoparticles, and the interaction between the base liquid and nanoparticles play an important role in determining the rheological behaviour of nanofluids. It should be noted that the non-Newtonian behaviour observed by Ding *et al* (2006) for carbon nanotube nanofluids, Kwak and Kim (2005) for CuO nanorod nanofluids, Tseng and Lin (2003) for concentrated aqueous suspensions of TiO₂, and Studart *et al* (2006) for Al₂O₃–MEK/T (methyl ethyl ketone/toluene) suspensions could be attributed, at least partially, to very different particle shape and concentration range. As this work focuses on spherical particles, particle shape effects will not be discussed further in this paper.

Figure 7 shows the measured viscosity at different temperatures for 0.5, 2.0 and 8.0 wt% nanofluids together with that for the pure EG. It can be seen that temperature has a strong effect on the viscosity of the nanofluids. Effort was therefore made to obtain a correlation between

Table 2. Empirical constants and deviation between equation (1) and the measurements.

Concentration	А	В	С	MaxD	MinD	AAD	GAD
0.0 wt%	-3.2114	0.86973	-154.57	+0.62%	-1.44%	-0.40%	0.78%
0.5 wt%	-3.1820	0.86285	-155.13	+0.31%	-1.58%	-0.41%	0.79%
1.0 wt%	-3.3289	0.91603	-150.35	+1.38%	-0.80%	+0.06%	0.59%
2.0 wt%	-3.5126	0.98375	-144.48	+0.75%	-1.31%	-0.19%	0.62%
4.0 wt%	-3.2517	0.91226	-150.74	+0.38%	-0.86%	+0.18%	0.63%
8.0 wt%	-3.7005	1.08082	-138.30	+0.26%	-1.65%	-0.26%	0.65%



Figure 8. Increase in the viscosity as a function of temperature.

the nanofluids viscosity (η) and the temperature. It was found that a similar expression to equation (1) (with η replacing η_0) describes the data well though the three empirical constants are different. Table 2 shows the values of these constants for all the nanofluids tested in this work. Also included in the table are deviations between the measured data and the empirical relation. It can be seen that deviation is less than 1.65%, indicating a very good representation of the experimental results by equation (1). If the nanofluids viscosity is expressed in the form of viscosity increase defined as $\eta_i = (\eta - \eta_0)/\eta_0$, figure 8 is obtained for all data. It is seen that the viscosity increase is a function of particle concentration but is (surprisingly) independent of temperature under the conditions of this work. The viscosity increase as a function of volume concentration is shown in figure 9 together with the prediction by the well-known Einstein equation (Einstein 1906, 1911) for dilute non-interacting suspensions of spherical particles, namely $\eta_i = \eta_r - 1 = 2.5\varphi$, where η_r is the so-called reduced viscosity given by $\eta_r = \eta/\eta_0$ and φ is the particle volume concentration. It can be seen that the nanofluids viscosity increases with increasing nanoparticle concentration in a nonlinear manner and the Einstein equation greatly underpredicts the nanofluid data although the equation has been shown to be able to predict the viscosity of suspensions for up to 10% by volume with an uncertainty less than $\sim 6\%$



Figure 9. Increase in viscosity as a function of nanoparticle concentration.

(Goodwin 2003). Regression of the experimental data gives the following correlation:

$$\eta_{\rm i} = \eta_{\rm r} - 1 = 10.6\varphi + (10.6\varphi)^2. \tag{2}$$

The binomial relationship between the viscosity increase and the nanoparticle volume concentration is similar to that for aqueous-based Al_2O_3 nanofluids (Das *et al* 2003; Wang *et al* 1999) and EG-based Al_2O_3 nanofluids (Wang *et al* 1999). It is different from the results of aqueous suspensions of spherical TiO₂ nanoparticles (He *et al* 2007) and PG based Al_2O_3 nanofluids (Prasher *et al* 2006), where linear correlations were obtained. For concentrated suspensions, the viscosity increase is shown to depend on nanoparticle volume concentration in an exponential manner; see for example Tseng and Lin (2003) for aqueous-based TiO₂ suspensions and Studart *et al* (2006) for MEK/T-based suspensions of Al_2O_3 . The reasons for these differences are discussed in section 4.

4. Analyses of the rheological data

4.1. Theoretical background

The theories of rheology of colloid dispersions have been well documented; see for example Russell *et al* (1991), Chow (1993), Petrie (1999), Goodwin *et al* (2000), Larson *et al* (2005) and Abdulagatov *et al* (2006). Investigation of the rheology of colloid dispersions started from Einstein's analysis of infinitely dilute suspensions of hard spheres (Einstein 1906, 1911). In his analyses, particles are assumed to be rigid, uncharged and without attractive forces; they are small enough so that the dilatational perturbation of the flow is unbounded and is able to decay to zero, and the disturbance around a particle does not interact with that around other particles. In such a case, a particle moves at the velocity of the streamline in line with the particle centre as shown in figure 10(a). The fluid passing the upper surface of the particle moves more rapidly than the particle known as the vorticity of the shear field and hence the following



Figure 10. Schematic illustrations of the interactions of particles with shear flow.

dispersion viscosity (η) :

$$\eta = \eta_0 \left(1 + [\eta] \varphi + O(\varphi^2) \right), \tag{3}$$

where $[\eta]$ is the intrinsic viscosity having a typical value of 2.5 for monodisperse dispersions of hard spheres. Equation (3) applies to $\varphi \leq 0.01$. When $\varphi \geq 0.01$, hydrodynamic interactions between particles become important as the disturbance of the fluid around one particle interacts with that around other particles. The viscosity in such a case is given by Batchelor (1977):

$$\eta(0) = \eta_0 \left(1 + 2.5\varphi + 6.2\varphi^2 + O(\varphi^3) \right), \tag{4a}$$

$$\eta(0) = \eta_0 \left(1 + 2.5\varphi + 7.6\varphi^2 + O(\varphi^3) \right), \tag{4b}$$

with equation (4*a*) for shear flow and (4*b*) for extensional flow, and $\eta(0)$ the so-called low shear limiting viscosity. Equations (4*a*) and (4*b*) are validated to $\varphi = 0.1$ for flows dominated by single particles (figure 10(a)) and pair-particle microstructures (figure 10(b)). They can be re-written in a general form:

$$\frac{\eta(0)}{\eta_0} = 1 + [\eta]\varphi + k_{\rm H} ([\eta]\varphi)^2 + O(\varphi^3), \tag{4c}$$

where $k_{\rm H}$ is the Huggin's coefficient and can be considered as the interaction parameter characterizing the colloidal interactions between particles as opposed to the purely hydrodynamic effect (Goodwin 2003). For $\varphi \gtrsim 0.1$, three-particle and multi-particle collisions (figure 10(c)) become increasingly important hence terms with an order of 3 and higher have to be considered for which there is no rigorous analysis. A semi-empirical relationship for the shear viscosity covering the full range of particle volume fraction was obtained by Krieger and Dougherty (1959):

$$\frac{\eta}{\eta_0} = \left(1 - \frac{\varphi}{\varphi_{\rm m}}\right)^{-[\eta]\varphi_{\rm m}},\tag{5}$$

where φ_m is the maximum particle packing fraction, which varies from 0.495 to 0.54 under quiescent conditions, and is approximately 0.605 at high shear rates. Inserting the φ_m value into equation (5) gives the following limiting viscosities at low and high shear conditions, $\eta(0)$ and $\eta(\infty)$, for monodisperse suspensions:

$$\frac{\eta(0)}{\eta_0} = \left(1 - \frac{\varphi}{\varphi_{\rm m}(0)}\right)^{-[\eta]\varphi_{\rm m}(0)} = \left(1 - \frac{\varphi}{0.5}\right)^{-1.25},\tag{6a}$$

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Figure 11. Relative viscosity as a function of volume fraction in the high- and low-shear limits.

$$\frac{\eta(\infty)}{\eta_0} = \left(1 - \frac{\varphi}{\varphi_{\rm m}(\infty)}\right)^{-[\eta]\varphi_{\rm m}(\infty)} = \left(1 - \frac{\varphi}{0.605}\right)^{-1.5125},\tag{6b}$$

where $\varphi_{\rm m}$ is taken as 0.5 at the low shear rate. Figure 11 plots the two limiting shear viscosities against volume fraction and four regions can be identified. In region A, dispersions behave as Newtonian fluids with no discernible shear dependence as $\eta(\infty)$ is almost equal to $\eta(0)$, whereas the shear-thinning behaviour may be observed in region B as $\eta(\infty) > \eta(0)$. The liquidlike viscoelastic behaviour is seen in region C with both the high and low shear viscosities accessible. In region C, $\eta(\infty)$ is much smaller than $\eta(0)$ so strong shear thinning is expected. The low shear viscosity is no longer accessible in region D and the behaviour is like that of a viscoelastic solid with a yield stress. However, the high shear viscosity is accessible and if the yield stress is exceeded, the structure can melt and flow can occur. Also shown in figure 11 are the results of the Einstein equation and equation (4*a*). Good agreement can be seen among the four equations for $\varphi < 0.1$. In fact, if φ approaches zero, the Krieger–Dougherty equation (5) reduces to the Einstein equation if a monomial expansion is performed, whereas a binomial expansion will lead to the Batchelor equation (4).

Having determined the two limiting shear viscosities above, the shear viscosity, η , as a function of shear rate can be determined from (Goodwin 2000):

$$\eta = \eta(\sigma_{\rm r}) = \eta(\infty) + \frac{(\eta(0) - \eta(\infty))}{1 + (\sigma_{\rm r}/\sigma_{\rm c})^n},\tag{7a}$$

$$\eta = \eta(Pe) = \eta(\infty) + \frac{(\eta(0) - \eta(\infty))}{1 + (Pe/Pe_c)^m},$$
(7b)

where σ_r is the reduced shear stress defined by $\sigma_r = \sigma a^3/k_B T$, σ is the shear stress which relates to the shear rate, $\dot{\gamma}$, according to $\sigma = \eta \dot{\gamma}$, a is the particle radius, k_B is the Boltzmann constant, T is the absolute temperature, m and n are power law indices (equal to 1 for monodisperse spherical particles), Pe is the Péclet number defined as $Pe = 6\pi a^3 \sigma/k_B T$, and the subscript 'c'

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Figure 12. Calculated shear thinning response for a hard sphere dispersion at $\varphi = 0.35$.

represents the characteristic point in the shear-dependent viscosity curve at which $Pe = \sim 1$. Pe = 1 implies physically that the diffusive and convective timescales are equal, which also gives a value of $\sigma_c \sim 0.053$ at the ambient temperature. Figure 12 shows a plot of equation (7*a*) with this value and n = 1 and 2/3 at $\varphi = 0.35$. Cross (1965) indicated that *m* would approach 2/3 with increasing polydispersity.

4.2. Analyses of the rheological behaviour of nanofluids

In this section, attempts are made to interpret the discrepancy in recent studies dedicated to the rheological behaviour of nanofluids. The high shear viscosity, shear thinning behaviour and temperature effect will be discussed and the discussion will be based mainly on the theoretical considerations presented in section 4.1.

4.2.1. High shear viscosity of nanofluids. Recent studies reveal that nanoparticles in nanofluids are mostly in the form of aggregates (He *et al* 2007; Kwak and Kim 2005; Prasher *et al* 2006a, 2006b, 2006c; Wen and Ding 2005). Assume that hydrodynamic forces are insufficient to break the network of particles in the aggregate down to individual primary particles, but only make the aggregates form spherical flow units with an effective volume fraction of φ_a , the viscosity in a high shear flow takes the following form according to equation (5):

$$\frac{\eta}{\eta_0} = \left(1 - \frac{\varphi_a}{\varphi_m(\infty)}\right)^{-[\eta]\varphi_m(\infty)},\tag{8}$$

with φ_a given by:

$$\varphi_{\rm a} = \frac{\varphi}{\varphi_{\rm ma}} \quad \text{and} \quad n_{\rm a} = \varphi_{\rm ma} \left(\frac{a_{\rm a}}{a}\right)^3,$$
(9)

where φ_{ma} is the maximum packing fraction of aggregates, a_a the radius of the aggregate, and n_a is the number of particles contained in an aggregate. As aggregates are often in the dendritic

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form and do not have a constant packing throughout the structure, it is assumed that the packing density in the aggregates decreases from centre to the edge following the following power law:

$$n_{\rm a} = \left(\frac{a_{\rm a}}{a}\right)^D. \tag{10}$$

Inserting equation (10) into (9) gives the effective volume fraction of the aggregates:

$$\varphi_{a} = \varphi \left(\frac{a_{a}}{a}\right)^{3-D}.$$
(11)

Combining equations (8)–(11) gives:

$$\frac{\eta}{\eta_0} = \left(1 - \frac{\varphi}{\varphi_{\rm m}(\infty)} \left(\frac{a_{\rm a}}{a}\right)^{3-D}\right)^{-[\eta]\varphi_{\rm m}(\infty)}.$$
(12)

The term *D* in the above equations is the so-called fractal index indicating the extent of changes in the packing fraction from the centre to the edge of the aggregates. Typical values of *D* are given in standard textbooks as $D \approx 1.8$ –2.5 for diffusion limited aggregation (DLA) and $D \approx 2.0$ –2.2 for rate limited aggregation (RLA); see for example Goodwin (2000). For nanofluids, the value of *D* is found to be 1.6–1.8 by both measurements and simulations (Wang *et al* 2003, Xuan *et al* 2003) and 1.8–2.3 by a thorough study of aggregation of nano-sized alumina suspensions (Waite *et al* 2001). If one takes *D* as 1.8 (Prasher *et al* 2006b), [η] as 2.5 and φ_m as 0.605 (Krieger and Dougherty 1959), equation (12) reduces to:

$$\frac{\eta(\varphi)}{\eta_0} = \left(1 - \frac{\varphi}{0.605} \left(\frac{a_{\rm a}}{a}\right)^{1.2}\right)^{-1.5125}.$$
(13)

Equation (13) is plotted in figure 13 in the form of η_i as a function of particle volume fraction for four effective radii of the aggregates of *a*, 3*a*, 3.34*a* and 4*a*, where the effective aggregate radius of 3.34*a* is obtained by fitting the experimental results of the current work to equation (13). This value is in fact consistent with the SEM analysis (figure 3), where the aggregates are seen in the size range of 70–100 nm and the primary nanoparticles have a diameter of ~25 nm. It is interesting to note that by expanding equation (13) according to the monomial and binominal equations with $a_a = 3.34a$ leads to $\eta_i = \eta(\varphi)/\eta_0 - 1 = \eta_r - 1 = 10.6\varphi + O(\varphi^2)$ and $\eta_i \approx 10.6\varphi + (10.6\varphi)^2 + O(\varphi^3)$, respectively. These two expressions are respectively equivalent to the correlation obtained by Prasher *et al* (2006d) and that obtained experimentally in this work (equation (2)). Equation (2) can also be re-written in a more general form:

$$\eta_{\rm i} = \eta_{\rm r} - 1 = [\eta]\varphi_{\rm a} + ([\eta]\varphi_{\rm a})^2 + O(\varphi_{\rm a}^3), \tag{14}$$

with $\varphi_a = \varphi (a_a/a)^{3-D}$ the effective volume fraction of aggregates. Equation (14) is consistent with (4c) for $k_H = 1$ and with the Einstein equation by monomial expansion for $a_a = a$.

Figure 14 shows a comparison between equation (14) for $a_a = 2a$, 3a and 4a with experimental data of this work and those reported in the literature (Das *et al* 2003a; He *et al* 2007; Prasher *et al* 2006d; Wang *et al* 1999). It can be seen that all the data points are bound by the curves for $a_a = 2a$ and $a_a = 4a$, and are very close to the curve for $a_a = 3a$. This suggests that the viscosity increase of nanofluids is mainly due to aggregation of nanoparticles and the effective size of the aggregates a_a is within 2–4 times the primary nanoparticles. The high shear viscosity of nanofluids increases with increasing effective size of aggregates for a given primary nanoparticle size. Figure 14 also shows that, given the effective radius of the aggregate,



Figure 13. Increment of viscosity as a function of volume fraction.



Figure 14. Increment of viscosities for spherical particle nanofluids.



(a) 0.01 vol. %



(b) 0.1 vol. %

Figure 15. SEM micrographs of nanofluids.

equation (14) provides an adequate prediction of the high shear viscosity of nanofluids by taking $[\eta] = 2.5$, D = 1.8 and $\varphi_m = 0.605$.

It is noted that the discrepancy between the measured data and equation (14) for $a_a = 3.34a$ is bigger at $\varphi_m = 0.001$ than that at higher concentrations (figure 13). This may be because the basic assumption of fractal aggregation is not applicable at very low concentrations at which the Einstein equation holds. This explanation is supported by SEM analyses; see figure 15, where aggregates for 0.01 and 0.1 vol% TiO₂ nanofluids are shown. It can be seen that nanoparticles are well dispersed at 0.01 vol%, whereas some aggregation occurs at 0.1 vol%. Considering the



Figure 16. Ratio of low to high shear viscosity as a function of volume fraction.

results for 0.2 vol% at which obvious aggregation occurs (figure 3), and high shear viscosity shown in figure 13, the lower bound for the applicability of equations (12)–(14) is estimated to be 0.1–0.2 vol%. This seems to be consistent with the experimental results of Kwak and Kim (2005) who found a big increase in viscosity at 0.2 vol%.

It is expected that there is also an upper concentration limit for equations (12)–(14) because simultaneous nucleation occurs with increasing particle concentration. This will lead to aggregates eventually touching each other and further growth of aggregates would require interpenetration so that the fractal aggregation is not possible. A rough estimation of the upper limit for $a_a = 3a$ and 4a with D = 1.8 gives respectively $\varphi_m = 0.162$ and 0.114 using equation (12). Therefore, equations (12)–(14) are less likely to be applicable to the concentrated dispersions that Tseng *et al* investigated (Tseng and Lin 2003; Tseng and Chen 2006).

4.2.2. Shear thinning behaviour. As mentioned before, some studies found Newtonian behaviour of nanofluids (Das *et al* 2003, 2006; Prasher *et al* 2006), while others observed non-Newtonian shear thinning behaviour (He *et al* 2007; Kwak and Kim 2005). Attempts are made in the following to interpret the discrepancy from three aspects, nanoparticle concentration, range of shear rate and base liquid properties.

As mentioned in section 4.1, dilute dispersions with $\varphi < 0.2$ should display no discernible shear dependence (region A in figure 11) although theoretically shear thinning behaviour exists for any liquid–solid dispersions. Nanofluids normally have a concentration below 0.1 so should not show shear thinning behaviour. However, due to aggregation as described in section 4.2.1, the effective volume fraction (φ_a) of nanofluids can be much higher than the actual solids volume fraction (φ); see equation (11). This can make the dispersion fall into regions B or even C in figure 11 hence the shear thinning behaviour. Figure 16 shows the ratio of low to high shear viscosities, $\eta(0)/\eta(\infty)$, as a function of actual solids volume fraction of particles in nanofluids containing aggregates with a size of $a_a = 2a$, 3a and 4a. It can be seen that the ratio $\eta(0)/\eta(\infty)$ becomes considerably larger than 1 when the volume fraction $\varphi_a \approx 0.2$. For the EG-based TiO₂



Figure 17. Viscosity of EG-based TiO₂ nanofluids as a function of shear rate.

nanofluids used in this work, if one takes $[\eta] = 2.5$, D = 1.8, $\varphi_m(0) = 0.5$ and $\varphi_m(\infty) = 0.605$, equations (6*a*) and (6*b*) become:

$$\frac{\eta(0)}{\eta_0} = \left(1 - \frac{\varphi_a}{\varphi_m(0)}\right)^{[\eta]\varphi_m(0)} = \left(1 - \frac{\varphi_a}{0.5}\right)^{-1.25},\tag{15a}$$

$$\frac{\eta(\infty)}{\eta_0} = \left(1 - \frac{\varphi_a}{\varphi_m(\infty)}\right)^{[\eta]\varphi_m(\infty)} = \left(1 - \frac{\varphi_a}{0.605}\right)^{-1.5125}.$$
(15b)

According to equation (7a), the relative viscosity of the nanofluid at any shear rate can be given as:

$$\frac{\eta}{\eta_0} = \frac{\eta(\sigma_{\rm r})}{\eta_0} = \frac{\eta(\infty)}{\eta_0} + \frac{(\eta(0)/\eta_0) - (\eta(\infty)/\eta_0)}{(1 + (\sigma_{\rm r}/\sigma_{\rm c})^n)}.$$
(16)

Considering the relationship between σ_r and σ , equation (16) becomes:

$$\frac{\eta}{\eta_0} = \frac{\eta(\sigma)}{\eta_0} = \frac{\eta(\infty)}{\eta_0} + \frac{(\eta(0)/\eta_0) - (\eta(\infty)/\eta_0)}{(1 + (\sigma/\sigma_c) \cdot (a_a^3/k_BT))},$$
(17)

for mono-dispersed nanofluids. Considering further $\sigma = \eta(\sigma)\dot{\gamma}$, figure 17 can be obtained, which plots the relative viscosity as a function of shear rate for EG-based TiO₂ nanofluids, where a_a is taken as 3.34*a*, temperature as 20 °C (293 K), and nanoparticle concentrations as 0.02, 0.04 and 0.08 by volume. Also included in figure 17 are the experimental data from the current work for a particle volume concentration of 0.018. Excellent agreement between the measured data and the prediction by equation (17) has been obtained. Figure 17 shows no obvious shear thinning behaviour of nanofluids containing nanoparticles up to a volume fraction of ~0.04, consistent with the data presented in section 3. Shear thinning behaviour could be seen for nanofluids with $\varphi = 0.08$ according to equation (17), which is consistent with the results shown in figure 16. Figure 17 also shows that the shear thinning behaviour only occurs at very low shear rates. For an EG-based TiO₂ nanofluid with $\varphi = 0.08$, the characteristic shear rate, $\dot{\gamma}_c$ is estimated at ~4 s⁻¹. This characteristic shear rate is far lower than that used in the reported



Figure 18. Characteristic shear rate as a function of viscosity of base liquid.

studies by, for example, Das *et al* (2003), which could be one of the reasons that the shear thinning behaviour of nanofluids was not observed.

The base liquid properties are expected to affect the rheological behaviour of nanofluids. Such an effect is reflected in the characteristic shear rate (shear rate at the characteristic point as shown in figure 12) expressed as:

$$\dot{\gamma}_{\rm c} = \frac{\sigma_{\rm c}}{\eta(\sigma_{\rm c})} \cdot \frac{k_{\rm B}T}{a_{\rm a}^3}.$$
(18)

As $\sigma_c \approx 0.053$, the characteristic shear rate $\dot{\gamma}_c$ is inversely proportional to the characteristic viscosity of nanofluids, $\eta(\sigma_c)$, and hence is a strong function of base liquid viscosity, η_0 , according to equations (15) and (17). Figure 18 shows the dependence of the characteristic shear rates for nanofluids on the base liquid viscosity for three cases, where nanoparticle size is taken as 25 nm and temperature as 293 K. The characteristic shear rate is seen to decrease sharply with increasing viscosity of base liquid particularly at relatively low viscosity. This indicates that a less viscous liquid like water will have a high characteristic shear rate in comparison with a highly viscous liquid like EG, PG and mineral oils. In other words, the shear thinning behaviour of nanofluids made from less viscous base liquids can be observed even at a relatively high characteristic shear rate. Take $a_a = 4a$, the typical characteristic shear rate for water-based nanofluids is 20–30 s⁻¹, which is much higher than that for EG-based nanofluids ($\sim 4 s^{-1}$). The characteristic shear rate for the water-based nanofluids agrees well with the experimental results for water-TiO₂ nanofluids (He *et al* 2007).

Figure 18 also shows the characteristic shear rate decreases with increasing volume fraction and/or increasing aggregate size, implying more concentrated suspensions with larger aggregates give a smaller characteristic shear rate. Such a phenomenon is because the ratio of particle motion due to the Brownian diffusion to that due to convection decreases with increasing viscosity, volume fraction or aggregate size. Figure 18 could be used as a guide to the shear rate dependence of the viscosity of nanofluids.

As a summary of the above discussion, for *dilute* nanofluids ($0 < \varphi \lesssim 0.001$), nanoparticles are well dispersed, the shear viscosity can be predicted by the Einstein equation, and there



Figure 19. Relative viscosity as a function of shear rate at different temperatures.

is no discernible shear-thinning behaviour. For *semi-dilute* nanofluids (~0.001 < $\varphi \lesssim 0.05$), nanoparticles aggregate, the viscosity can be predicted by equations (12)–(14), and there is no obvious shear-thinning behaviour. For *semi-concentrated* nanofluids (~0.05 < $\varphi \lesssim 0.10$), nanoparticles aggregate, the shear viscosity can be predicted by equations (12)–(14), and there is a clear shear-thinning behaviour. Suspensions with $\varphi \gtrsim 0.10$ are *concentrated*, interpenetration of aggregation may occur and they are out of the normal range of nanofluids.

4.2.3. Temperature effect. Temperature has an effect on both high shear viscosity and shear thinning behaviour of nanofluids. The effect of temperature on the high shear viscosity of nanofluids is reflected in equation (15*b*), which states that the relative high shear viscosity of nanofluids is the function of three variables, the maximum volume fraction φ_m , the effective volume fraction φ_a and the intrinsic viscosity [η]. For a given nanofluid at temperatures not far from the ambient temperature, the three variables are independent of temperature. This explains the temperature independence of the relative shear viscosity discussed in section 3. Physically, the temperature-independent behaviour is due to negligible Brownian diffusion compared with convection in high shear flows. In addition, for dilute nanofluids with $\varphi < 0.05$ investigated in this work, the second terms in the brackets of the right-hand side of equations (15*a*) and (15*b*) are vanishingly small so $\eta(0)$ is approximately equal to $\eta(\infty)$, hence the temperature dependence, if any, is very weak. However, for semi-concentrated nanofluids with $0.05 < \varphi \leq 0.1$, the shear thinning behaviour can be obvious as the second terms in the brackets of the right-hand side of equations (15*a*) and (15*b*) differs significantly.

The temperature effect on the shear thinning behaviour of nanofluids can be analysed by looking at the dependence of the characteristic shear rate, $\dot{\gamma}_c$, on temperature using equations (17) and (18). Figure 19 shows such an analysis, which plots the relative viscosity of EG-based TiO₂ nanofluids for $\varphi = 0.08$ and $a_a = 3.34a$ as a function of shear rate for three different temperatures. It can be seen that the characteristic shear rate increases with increasing temperature, implying a stronger shear thinning behaviour at high temperatures. Physically, this is because the Brownian diffusion becomes stronger at a higher temperature, hence *Pe* decreases and the characteristic shear rate moves to the right in figure 19.

5. Conclusions

Both experimental work and theoretical analyses are performed with aims of more fundamental understanding of the rheological behaviour of nanofluids and interpretation of the discrepancy in the reported rheological behaviour of nanofluids in the recent literature. The experiments are carried out on EG-based TiO₂ nanofluids containing 0.5, 1.0, 2.0, 4.0 and 8.0% by weight spherical nanoparticles at 20–60 °C. The theoretical analyses are performed on the high shear viscosity, shear thinning behaviour and temperature dependence. The following conclusions are obtained.

- 1. EG-based nanofluids show Newtonian behaviour under the conditions of this work with shear viscosity as a strong function of temperature and particle concentration. The relative viscosity depends on particle concentration in a nonlinear manner but is independent of temperature.
- 2. The high shear viscosity of nanofluids can be predicted by the Krieger–Dougherty equation if the solids volume fraction is replaced by the volume fraction of nanoparticle aggregates. For spherical nanoparticles, an aggregate size of approximately 3 times the primary nanoparticle size gives the best prediction of the experimental data of both this work and those reported in the literature.
- 3. The non-Newtonian shear thinning behaviour of nanofluids is determined mainly by three parameters, the effective particle volume fraction, the range of shear rate and the viscosity of the base liquid and can be characterized by the characteristic shear rate. The characteristic shear rate decreases with increasing volume fraction, increasing base liquid viscosity, or increasing aggregate size because the ratio of particle diffusion due to Brownian motion to that due to convection decreases with an increase in the three parameters. These findings explain the reported controversy of the rheological behaviour of nanofluids in the literature.
- 4. At temperatures not very far from room temperature, the relative high shear viscosity is independent of temperature due to negligible Brownian diffusion in comparison to convection at high shear flows, in agreement with the experimental results. However, the characteristic shear rate can have strong temperature dependence, thus affecting the shear thinning behaviour of nanofluids.
- 5. The rheological behaviour of nanofluids could be categorized into four groups: (i) *dilute* nanofluids ($0 < \varphi \leq 0.001$) with well dispersed nanoparticles, the shear viscosity fits the Einstein equation and there is no discernible shear-thinning behaviour; (ii) *semi-dilute* nanofluids ($\sim 0.001 < \varphi \leq 0.05$) with aggregation of nanoparticles, the shear viscosity fits the modified Krieger–Dougherty equation and there is no obvious shear-thinning behaviour; (iii) *semi-concentrated* nanofluids ($\sim 0.05 < \varphi \leq 0.1$) with aggregation of nanoparticles, the shear viscosity fits the modified Krieger–Dougherty equation and there is obvious shear-thinning behaviour; (iii) semi-concentrated nanofluids ($\sim 0.05 < \varphi \leq 0.1$) with aggregation of nanoparticles, the shear viscosity fits the modified Krieger–Dougherty equation and there is obvious shear-thinning behaviour; and (iv) *concentrated* nanofluids ($\varphi \geq 0.10$) with interpenetration of aggregation, this is out of the normal concentration range of nanofluids.

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