# TURBULENCE MODULATION IN PIPE FLOWS OF DISPERSED AND AGGREGATED COLLOIDAL SUSPENSIONS

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#### ABSTRACT

The streamwise component of turbulence intensity is studied in a 26mm NB horizontal pipe loop. Colloidal silica suspensions were prepared in 10<sup>-4</sup>M and 1M KNO<sub>3</sub> electrolytes providing two contrasting colloidal suspensions; i) silica particles remain dispersed in the presence of a strong electrical double layer force, and ii) the repulsive component of interaction is sufficiently diminished such that collisions between particles lead to the formation of particle clusters (aggregates). Changes in the turbulence characteristics of the two colloidal suspensions are compared with the turbulence characteristics of a sedimentfree flow. Little change (within error) in the suspension's turbulence characteristics over a range of Reynolds numbers (3,000-10,000) is observed between the dispersed colloidal suspension and the sediment-free flow. An increase in the electrolyte concentration for equivalent solids loading (~5.5-5.7% vol.) leads to a slightly different behaviour with two critical Reynolds numbers identified. The first critical value (Re = 5,500) is observed when the RMS values for the aggregated suspension exceed the RMS values for the dispersed suspension and the sediment-free flow. The second critical Reynolds number (Re  $\approx$  8,000) is identified when the RMS profile for the aggregated suspension converges towards the RMS profiles for the dispersed and sediment-free flow. The critical Reynolds numbers are associated to turbulence modulation in the presence of aggregates and the break-up of aggregates once a critical fluid shear stress is exceeded.

#### INTRODUCTION

Solid-liquid suspensions are ubiquitous in the process industries, and it is therefore unsurprising that the flow properties of such systems are extensively studied. In recent years a topic of great interest has been the modulation of fluid turbulence in the presence of suspended particles. Researchers have shown that the addition of particles to a fluid can either act to enhance or dampen the fluid turbulence. An early review of turbulence modulation was published by Gore and Crowe (1989) who compared 15 experimental data sets, accounting for a variety of geometries and flow conditions. By comparing the turbulence intensity as a function of the particle diameter (d<sub>p</sub>) and the characteristic length of the most energetic eddies (le), the authors concluded that there is a critical length scale  $(d_p/l_e)$  where the transition from turbulence attenuation to augmentation, or vice versa, is observed. This length scale is given to be approximately 0.1. Greater than 0.1, turbulence augmentation is generally associated

<sup>†</sup>Current address: The Energy Institute, Steinman Hall, City College New York, 140th St. at Convent Ave., NY 10031 with secondary flow (vortex shedding) which acts to increase the turbulence intensity of the fluid, whilst turbulence attenuation ( $d_p/l_e < 0.1$ ) is often observed during the transportation of fine particles; a result of work done by the turbulent eddies. The Gore and Crowe (1989) publication provides a reasonably high level assessment of a subject that is still not well understood.

In reviewing the literature many parameters that can influence the turbulence characteristics of a solid-liquid system have been extensively studied both experimentally and theoretically. However, these studies have a common theme, in that the particles remain dispersed during transportation. To the authors' knowledge, the effect of particle clusters on fluid turbulence has received very little attention to date (Hagiwara et al., 2002). This lack of work is surprising given that the Gore and Crowe (1989) publication highlighted the omission of the Maeda et al. (1980) data set which displayed an enhancement in the fluid turbulence when the critical length scale is less than 0.1. This deviation from the trend was suggested to be related to the formation of particle clusters in the fluid, which generate additional fluctuations not considered when the particles remain dispersed. The focus of the current study is to investigate these flow systems in further detail.

Particle clusters (aggregates) are formed when the solids become surface active. For example, static charging in powder flow or ionisation when a solid is dispersed in an aqueous electrolyte solution can lead to the formation of aggregates. In an aqueous electrolyte environment very fine particles (colloids) exhibit extremely low inertial forces; as a result the surface charge plays a significant role in their interaction, partially governing the size and shape of the Open porous aggregates are formed under aggregate. conditions what are commonly referred to as diffusionlimited cluster-cluster aggregation, DLCA (Franks et al., 2004). DLCA occurs when the dispersive component for particle stability is negligible, leading to a situation where all collisions result in a sticking event producing large open porous clusters which are unlikely to undergo rearrangement to produce more compact clusters. With a small potential barrier to aggregation the probability of a sticking event when the particles come into close proximity is reduced. The aggregation kinetics are suitably described by the reaction-limited, cluster-cluster aggregation, RLCA, model (Franks et al., 2004), where the particles have an opportunity to diffuse into the core of the aggregate before they become attached. This mechanism, which is limited by the reaction of the particle to the cluster particles, produces smaller more compact aggregate structures (Figure 1).

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Figure 1. Simulated aggregate structures formed under the DLCA mechanism (a) and RLCA mechanism (b). Image taken from (Biggs, 2006) with simulations completed by Dr. Graeme Bushell, Department of Chemical Engineering, UNSW.

The interaction potential between colloidal particles is suitably described by the DLVO (Derjaguin and Landau, 1941, and Verwey and Overbeek, 1948) theory, and is given as the summation of the attractive van der Waals potential and the electrical double layer repulsive potential. When two like surfaces approach, there is a like-like charge repulsion effect which prevents the surfaces from intimate contact. However, the surface charge can be effectively "screened" by increasing the ionic concentration of the solution, which allows the surfaces to approach to within a shorter separation distance before the surfaces "sense" each other and begin to repel. As the ionic concentration of the suspension increases, the thermal energy barrier (kT)preventing interaction decreases, and a condition can be met where the kinetic energy of the particles (Brownian diffusion, gravitational sedimentation, fluid shear) is sufficient to drive the surfaces into contact. Upon contact, the surfaces are likely to "stick" forming aggregates which can be several orders of magnitude greater in size with respect to the primary particle.

Figure 2 illustrates the change in the total interaction energy (V<sub>T</sub>) as a function of the separation distance and electrolyte concentration. An increase in the electrolyte concentration lowers the energy of interaction to the point where the suspension is un-stable, and the system is energetically more favourable to form aggregates, e.g. at  $2 \times 10^{-1}$ M - separation distance less than 10nm.

With improvements in attrition technologies and the advancement of "bottom-up" engineering applications, colloidal particles and their handling is becoming increasingly more important in the process industries. In this study we aim to highlight the importance of solution chemistry when transporting fine particulate solids.



Figure 2. Total potential energy of interaction curves as a function of electrolyte concentration.

### **EXPERIMENTAL WORK**

#### Materials

An ultra high purity (> 99.5% SiO<sub>2</sub>), near monodisperse, silica sample (SP-1B) was obtained from Fuso Chemical Co., Ltd. Osaka, Japan (Figure 3). This silica sample had a density of  $2.26g/\text{cm}^3$  (determined using a Micromeritics AccuPyc 1330 Pycnometer), and a mean particle diameter of  $0.79\mu\text{m}$ , measured using a centrifugal sedimentation particle size analyser supplied by CPS Instruments Europe. All chemicals and other reagents were of analytical grade and were supplied by Aldrich Chemical Company, Inc. (United Kingdom). The silica particles were dispersed in KNO<sub>3</sub> electrolyte solutions (1M or  $10^{-4}\text{M}$ ), and all pH adjustments were made using complementary acid (HNO<sub>3</sub>) and base (KOH). All water used throughout this study was Milli-Q<sup>®</sup> grade water with a conductivity of approximately 0.05 µs/cm.



Figure 3. Fuso silica spheres and particle size distribution.

#### Method

The fluid turbulence in fully developed pipe flow was studied using ultrasonic Doppler velocity profiling (UDVP). An ultrasonic probe positioned at 45° to the flow axis and submerged 1mm into the flow stream was used to collect 512 velocity profiles over a 30 second period. Flow profiles were collected at different flow rates between  $7.5 \times 10^{-1}$  m<sup>3</sup> hr<sup>-1</sup> and  $5 \times 10^{-2}$  m<sup>3</sup> hr<sup>-1</sup>, with each measurement collected after 1 minute of steady-state flow. The fluid turbulence was determined from the root-mean square values (*u*') of the streamwise component of the flow, where the RMS is given as:

$$u' = \left[\frac{1}{n}\sum_{i=1}^{n} (u-U)^2\right]^{0.5}$$
(1)

where u is the instantaneous velocity and U the mean streamwise velocity.

## **RESULTS AND DISCUSSION**

Comparing the turbulence intensities (turbulence intensity = RMS/depth-averaged streamwise velocity) of the aggregated and dispersed suspensions at an equivalent solids concentration (see Figure 4), there is a clear enhancement in the near-wall turbulence when the suspension is aggregated. With the edge of the buffer layer at r/R = 0.82 (Re = 5,200) and r/R = 0.87 (Re = 7,500) the turbulence modulation behaviour extends into the mainstream region of the flow. Considering the buffer region first, the turbulence intensity will be a function of the fluid-wall, particle-fluid, particle-wall and particle-particle interactions. Fluid-wall interactions relate to the intermittent low-speed fluid ejections from the wall (fluid bursts) and high-speed fluid injections towards the wall (fluid sweeps). With a particle relaxation time ( $\tau_p$  =  $\rho_{\rm p} d_{\rm p}^{2}/18\mu$ ; Stokes regime) smaller than the characteristic time of the energy containing eddies ( $t_e = \lambda u''$ ), the particles will follow the flow, neither dampening or enhancing the intensity of the sweeps or bursts in the fluid boundary layer. The particle Reynolds numbers for the primary particle and aggregate are  $3.5 \times 10^{-7}$  and  $6.7 \times 10^{-5}$ , respectively. With such low particle Reynolds numbers the colloidal particles and aggregates in suspension should not influence the fluidwall or particle-fluid interactions. However, at such a high solids concentration cumulative effects may result where small interferences between the fluid and particle, which are commonly considered to be negligible at extremely low solids concentrations, may in fact become significant and modulate the turbulence intensity. For example, particle collisions at the pipe wall and with each other generate small scale fluctuations within the fluid which in dilute flows can be considered negligible, while in concentrated flows the effect may become measurable.

The differences in the buffer layer turbulence intensities may also result from small differences in the near-wall velocity gradients. With the fluid shear stress proportional to the velocity gradient, an increase in the velocity gradient provides a more favourable environment for eddy production and additional turbulence. Comparing the buffer layer velocity gradients (Table 1):  $du_x/dy 1M \text{ KNO}_3 > du_x/dy 10^{-4}M \text{ KNO}_3$  and  $du_x/dy$  water.

Table 1. Velocity gradients in the near wall region  $y^+ < 30$ . Units:  $u_x$  mm/s, r/R(y) mm, Re = 7,500.

	du <sub>x</sub> /dy
1M KNO	-75.4
10 <sup>-4</sup> M KNO <sub>3</sub>	-61.7
Sediment free	-66.4



Figure 4. Streamwise turbulence intensities. Symbols: square -1M KNO<sub>3</sub> (5.7% vol.), circle  $-10^{-4}M$  KNO<sub>3</sub> (5.5% vol.), solid triangle – water. a) Re = 7,800, b) Re = 5,200.

Centre-line variations in the turbulence intensities are observed in the raw data profiles, and have been summarised in Figure 5 illustrating the percentage change in turbulence intensity when compared to a sediment-free flow. An average of 10 individual measurements at each velocity provides a suitable assessment of the error of The turbulence intensities of a dispersed uncertainty. suspension do not appear to deviate too much from those measured in a sediment-free flow. The transition from turbulence augmentation to attenuation as the mean flow velocity is increased may not be a real effect, and can be considered within experimental error. However, the data does suggest to a certain degree that the centre-line turbulence intensities are enhanced in the presence of an aggregated suspension. At the wall where the enhanced turbulence can be accounted for by particle-wall interactions, the centre-line turbulence intensity can only be particle-fluid and particle-particle influenced by interactions. Typically, the centre-line turbulence intensities are influenced by vortex shedding from wakes behind particles (Achenbach, 1974: Gore and Crowe, 1989: Kim et al., 2005) and changes in the fluid viscosity (Zisselmar and Molerus, 1979). In this study, interpretation of the experimental data and critical analysis of the observed behaviour is difficult with no comparable data sets available to validate such behaviour. Commonly, when the particle length scales are significantly smaller than the Kolmogoroff length scale, relatively small changes in the turbulence intensities are observed which are frequently associated with a change in the suspension viscosity.

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Figure 5. Percentage change in centre-line turbulence intensity relative to water: open symbols  $-1M \text{ KNO}_3$  (5.7% vol.), closed symbols  $-10^{-4}M \text{ KNO}_3$  (5.5% vol.), shaded region – water.

Possible reasons for the observed enhancement in the centre-line turbulence intensities can be related to variations in the micro-hydrodynamic disturbances at the solid-liquid interface, which can result from; inter floc flow fields in an open porous structure (Yang et al., 2007), changes in the hydrodynamic drag or changes in the angular rotation rates of the particles. Because of the concentrated nature of the suspensions examined, which precluded the use of PIV, information on particle rotation and slip velocities could not be collected. Figure 6 shows a clear difference in the particle size and shape in an aggregated suspension which may lead to the development of complex flow fields and enhanced fluid disturbances.

Figures 7 (a) and (b) compare the RMS values of the streamwise component of the single phase fluid, dispersed and aggregated suspensions, and an additional dispersed suspension containing silica spheres with a mean particle diameter equal to  $27\mu m$  (Spheriglass 2000). All particle-laden suspensions are of equivalent solids loading. Both figures illustrate the changes in the RMS values as the flow progressively develops into fully developed turbulence. The distinguishing sharp peak in the flow fluctuations around the transition region is attributed to intermittent turbulence in the form of turbulence puffs (Wygnanski and Champagne, 1973; Wygnanski et al., 1974).

Above a critical Reynolds number (Re  $\approx$  5,500) the RMS streamwise component for an aggregated suspension exceeds the RMS values measured in a sediment-free flow and dispersed suspension flow, with the increase corresponding to the enhancement in turbulence intensity. The enhancement in the RMS values of the aggregated suspension correlate with the enhancement in the RMS values for the dispersed suspension containing 27µm silica spheres. The larger silica spheres which interact more with the fluid (vortex shedding) appear to modulate the turbulence to the same extent as the smaller aggregates. As the Reynolds number exceeds Re  $\approx$  8,000 the RMS profile for the aggregated suspension begins to converge with the dispersed suspension and sediment-free profiles. Such behaviour is in agreement with the thought that a critical

fluid shear stress is reached beyond which the aggregates undergo gradual break-up.



Figure 6. Primary particles in the dispersed phase  $(10^{-4}M \text{ KNO}_3)$  and bound together in the aggregated phase (1M KNO<sub>3</sub>). Images collected using Sysmex Flow Particle Image Analyser (FPIA-3000). Singlets and doublets imaged at low concentration electrolytes, with multi-particle clusters imaged at high electrolyte concentrations.



Figure 7. Root mean square values: a) solid square  $-10^{-4}$ M KNO3 (5.5% vol.), crossed square  $-10^{-4}$ M KNO<sub>3</sub> (12.7% vol.), open square -1M KNO<sub>3</sub> (5.7%vol), circle - water; b) symbols as in (a), triangle  $-27\mu$ m silica spheres (5.5% vol.).

Further validation for aggregate break-up is provided in the analysis of a flow curve obtained using a Bohlin CVO-R rheometer, shown in Figure 8. With an increase in shear rate ( $\phi$ ) the suspension viscosity ( $\eta$ ) in the aggregated suspension gradually decreases (shear thinning) until both the aggregated and dispersed flow curves overlap. A reduction in the suspension viscosity represents a lowering in the resistance to flow, which is common when particles align themselves within the flow field or undergo structure break-down. Comparing the shear rate versus viscosity profile and the pipeline pseudo-shear rate (8V/D) versus RMS profile (Figure 7a.), the critical shear rate when the aggregated and dispersed suspensions have similar properties (viscosity and RMS values) corresponds within reasonable agreement.

0.006 0.004 1 (Pa.s) □0 □0 0.002 0.000 100 200 300 400 500  $\gamma(s^{-1})$ 

Figure 8. Flow curve, viscosity versus shear rate: square  $-10^{-4}$ M KNO<sub>3</sub> (5.5% vol.), circle -1M KNO<sub>3</sub> (5.7% vol.).

### CONCLUSIONS

The data shows that the turbulence properties of concentrated colloidal suspensions can be modulated through inducing particle aggregation in the suspension. It should be acknowledged that the percentage change in the turbulence intensities is relatively small in comparison with published data; such small changes are a result of the very fine particulates used in the current study. To the authors' knowledge, this is the first study where the turbulence properties of colloidal suspensions have been investigated. Both the near-wall region and centre-line turbulence intensities are increased in the presence of aggregates, while the dispersed particles have minimal affect on the level of fluid turbulence. A comparison of the streamwise velocity fluctuations for sediment-free and particle-laden flows (dispersed and aggregated) identified an initial critical Reynolds number where the presence of aggregates enhances the turbulence intensity when compared with a dispersed suspension and a sediment free flow. A second critical Reynolds number (higher flow velocity) is identified when the aggregates begin to break-up as the particleparticle contact strength is exceeded by the fluid shear, and the RMS profile of the aggregated suspension begins to converge with the RMS profiles of the dispersed colloidal suspension and sediment free flow.

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