Chapter 1

Introduction

Granular materials and their suspension in liquids are prevalent in a wide range of natural and man-made processes. These include the industrial handling of seeds and slurries, clogging of drilling wells, and geological phenomena such as landslides and debris flows. Because of the complexity of having more than one phase (the solid and the fluid one), most of the understanding of how these materials flow is based on empirical observations, hampering, for example, the design of efficient transport of a suspension of solids in a fluid medium. Therefore the goal of this research is to help develop constitutive models that predict how liquid-solid mixtures behave when sheared as a function of various physical parameters, using carefully controlled experiments to validate and refine such models. The work presented in this thesis focuses on liquid-solid mixtures, and unlike the mechanics of dry granular material flows which are dominated by collisions and friction, the mechanics for these mixtures involve the interaction between the solid particles, the inertial effects from both liquid and solid phase, and viscous effects of the liquid. In particular, the effects of particle concentration and the density ratio between the two phases are studied under shear conditions where particle collisions might become important. A review of previous rheological experiments and the key parameters that govern the behavior of liquid-solid mixtures is presented.

1.1 Rheology of non-inertial suspensions

There is an extensive work done in the rheology of suspensions; however, most of these studies cover mixtures with low Reynolds number (Re) (from 10^{-6} to 10^{-3}), where Re is defined as $Re = \rho\dot{\gamma}d^2/\mu$, ρ and μ are the density and dynamic viscosity of the suspending liquid, $\dot{\gamma}$, is the shear rate and d is the particle diameter. Rutgers (1962) and Barnes (1989) did a summary of earlier studies where emphasis was made on the relationship between the particle concentration and the effective relative viscosity (μ'). The latter is defined as the proportionally function between the shear stress (τ) and shear rate of the mixture assuming that the mixture is Newtonian,

$$\tau = \mu' \dot{\gamma}$$

Rutgers (1962) examined the dependence of the relative viscosity on concentration for rigid spheres of more or less monodisperse character. He observed that with good dispersion, no structure-formation, no adsorption or solvation, and no electrical disturbance, the relation between the relative viscosity and the volume fraction may be independent of sphere size, in the size range of $0.264\mu m$ to $177\mu m$. Three regimes were recognized: (i) a dilute regime for volume fractions (ϕ) less than 0.02, where the relative viscosity (μ'/μ) depends almost linearly on ϕ and the dilute suspension exhibits a Newtonian behavior; (ii) a semi-dilute regime for $\phi \leq 0.25$, where μ'/μ exhibits a higher dependance on the particle concentration but the suspension behavior is still approximately Newtonian; (iii) a concentrated regime for $\phi \geq 0.25$, where μ'/μ increases rapidly with volume fraction and exhibits a shear thinning behavior. These studies cover a range of vanishingly small Re numbers only ($1.2 \times 10^{-6} \leq Re \leq 2 \times 10^{-5}$).

Effective viscosity models

The first person to address theoretically the suspension behavior in the dilute limit was Einstein (1906). Based on the hydrodynamics around a single sphere, Einstein derived the relative viscosity of such dilute suspension:

$$\frac{\mu'}{\mu}(\phi) = 1 + B\phi$$

where B is often referred as Einstein coefficient or 'intrinsic viscosity' and its value has not been validated (Mueller et al., 2010). Numerous expressions have been proposed to extend the range of validity of Einsteins expression to higher concentrations (Cheng and Law, 2003). They are either theoretical expansions or empirical expressions. The theoretical expansions are usually expressed in the form of power series:

$$\frac{\mu'}{\mu}(\phi) = 1 + k_1\phi + k_2\phi^2 + k_3\phi^3 + ...$$

This relation does not hold for volume fractions higher than 0.25 and even for $\phi < 0.25$, the polynomial relationship describe experimental data poorly (Rutgers, 1962; Thomas, 1965; Barnes, 1989; Mueller et al., 2010; Abedian and Kachanov, 2010). One reason for this is that the polynomial relation predicts a finite value of the relative viscosity for solid fractions close to one, which is physically impossible given that the maximum volume fraction for spherical particles of the same diameter is $\phi_m \approx 0.74$ (hexagonally close-packed arrangement). At this high solid fraction the relative viscosity must be infinite. For particles that are randomly distributed, the densest packing (random close packing) obtained is lower. Experimentally, the value of the random close packing ranges between

0.627 and 0.64 (Scott, 1960; Haughey and Beveridge, 1966; Scott and Kilgour, 1969); numerically, ϕ_m has been reported to be between 0.61 and 0.648 (Finney, 1970; Bennett, 1972; LeFevre, 1973; Torquato et al., 2000; O'Hern et al., 2002). Physically, ϕ_m represents the limiting fraction above which flow is no longer possible. However, higher packing is observed for particles that had been sheared (Rutgers, 1962; Tsai and Gollub, 2004). This suggests that shearing imposes additional structure to the particles distribution, and for high shear rates the particles are at volume fractions higher than the ϕ_m found for zero and moderate shear rates (der Werff and de Kruif, 1989). The model that best fits the experimental data for non-Brownian suspensions at higher solid fractions includes ϕ_m as a parameter. Considering a suspension with uniformly distributed particles, Krieger and Dougherty (1959) expanded Einstein's equation to higher concentration. Their equation relates the relative viscosity with ϕ_m as follows:

$$\frac{\mu'}{\mu}(\phi) = \left(1 - \frac{\phi}{\phi_m}\right)^{-C\phi_m}.$$

This equation tends to Einstein's equation when ϕ tends to zero and it has been used widely to fit experimental data (Jeffrey and Acrivos, 1976; Pabst, 2004; Pabst et al., 2006; Mueller et al., 2010), where C and ϕ_m are used as fitting parameters. Different theoretical and empirical correlations for the relative viscosity have been proposed with the same functional form as the Krieger-Dougherty equation (Maron and Pierce, 1956; Jeffrey and Acrivos, 1976; Quemada, 1977; Leighton and Acrivos, 1987a,b) .

Zarraga et al. (1999) studied the total stress of concentrated non-Brownian suspensions of spheres (43 μm glass beads) in Newtonian fluids. Using three different geometries (rotating rod, parallel plate, and cone and plate measurements), they measured the relative viscosity and proposed an empirical model in the form of Krieger-Dougherty equation:

$$\frac{\mu'}{\mu}(\phi) = \frac{\exp(-2.34\phi)}{\left(1 - \frac{\phi}{\phi_m}\right)^3},$$

where $\phi_m = 62\%$ for the particles used in their experiments. Their empirical model is in good agreement with other non-Brownian suspensions (Acrivos et al., 1993; Ovarlez et al., 2006; Bonnoit et al., 2010) for a Reynolds number range of 1×10^{-6} to 3×10^{-2}. Similarly, exponential formulas for computing the relative viscosity of the form

$$\frac{\mu'}{\mu}(\phi) = exp\left(\frac{D\phi}{1 - \phi/\phi_m}\right)$$

have been developed both theoretically and empirically, where D and ϕ_m are used as fitting parameters(Vand, 1948; Mooney, 1951; Cheng and Law, 2003).

All of these formulations are done under the assumption that the effective viscosity of the suspension is only a function of particle concentration. Other approaches on how to deal with the shear viscosity on concentrated suspensions include energy dissipation in the space between particles (Frankel and Acrivos, 1967; Hoffmnn and Kevelam, 1999), and the re-organization of particles in plane sheets that move one on top of another (Hoffmann, 1972).

Non-Newtonian behavior for concentrated non-inertial suspensions

Non-Newtonian behaviors had been observed in semi-dilute and concentrated suspensions at low Reynolds number. The shear thickening behavior was well reviewed by Barnes (1989) and it was found that there is a wide variety of suspensions that show a shear thickening behavior. Barnes' observations suggest that given the right circumstances, *all* suspensions of solid particles will show the phenomenon. Nevertheless, Barnes' review only covered particle size smaller than $100\mu m$, where the Brownian motion is present (Stickel and Powell, 2005). Evidence of the presence of a yield stress and shear-thinning behavior has also been extensively reported (Rutgers, 1962; Acrivos et al., 1994). Acrivos et al. (1994) studied the quasi-static regime of a suspension of rigid, non-colloidal particles immersed in a Newtonian fluid. They found a shear thinning behavior even for values of the solid fraction as low as 0.20. They explained such observations by means of particle distributions, which occurred due to a slight mismatch in the densities of the two phases.

Heymann et al. (2002) found the presence of a yield stress that was not single value for high volume fractions. This suggested that at high concentrations there is an elasto-viscous region where the particles form a network that deforms elastically at low shear rates and breaks up when the yield stress is reached.

Another factor that can influence the behavior of the suspension is Brownian motion: irregular motion of particles suspending in a fluid due to their colliding, thermally excited atoms and molecules. At room temperature and when particles are smaller than 100 μm, Brownian motion is present. The presence of Brownian motion is dictated by the Péclet number (Pe), which relates the rate of advection of a flow to its thermal diffusion and is defined as:

$$Pe = \frac{6\pi\mu d^3\dot{\gamma}}{kT},$$

where k is the Boltzmann constant and T is the absolute temperature. Suspensions with $Pe > 10^3$ are considered non-Brownian (Stickel and Powell, 2005). The present work deals with liquid-solid mixtures with Péclet numbers higher than 10^{10} where the effects from Brownian motion are negligible.

1.2 Rheology of inertial suspensions

The studies described in Section 1.1 were done in the limit of zero Reynolds number. Most of the work done on suspensions are often done in this limit (Brady and Bossis, 1988; Ladd, 1994; Zarraga et al., 1999; Brady, 2001; Sierou and Brady, 2002; Brady et al., 2006). Non-inertial suspensions represent one extreme of the studies of particulate flows. On the other extreme are studies of granular flows, which, unlike non-inertial suspensions where the particle inertia is negligible and the mechanics of the flow are dominated by hydrodynamic forces, the momentum transfer in granular flows is governed by particle collisions, and the interstitial fluid interaction with the particles is generally assumed to be negligible. Studies of granular flows have also been done extensively (Forterre and Pouliquen (2008) presents a review on such studies). However, there are fewer studies between these two extremes where both the inertia of one or both phases and the viscous effects of the fluid are important.

Stickel and Powell (2005) summarized the different types of non-Newtonian behavior observed in suspensions in terms of the effects of Brownian motion and the effects of inertia. Stickel and Powell (2005) considered that at steady state, the viscosity of a suspension is a function of 5 dimensionless numbers:

$$\frac{\mu'}{\mu} = f(\phi, Pe, Re, St, Ar),$$

where St is the Stokes number defined as:

$$St = \frac{1}{9}\frac{\rho_p}{\rho_f}Re,$$

and Ar is the Archimedes number which describes the ratio of gravitational forces to viscous forces and it is defined as:

$$Ar = \frac{gd^3\rho|\rho_p - \rho|}{\mu^2}.$$

Based on dimensionless analysis, Stickel and Powell (2005) constructed a "phase diagram", where the suspension may be expected to behave as a Newtonian fluid for large ranges of Pe and vanishingly small Re number, shear thickening for large Pe and Re, and shear thinning for both Pe and Re vanishingly small, as depicted in Figure 1.1. For non-Brownian systems (Pe$\rightarrow\infty$) the Peclet number can be neglected; therefore, the relative viscosity is a function of only the volume fraction, Archimedes, Stokes and Reynolds number ($\mu'/\mu = f(\phi, Re, St, Ar)$). For systems where the density of the two phases match, and considering a steady-state, the density ratio can be neglected and consequently so can the Stokes and Archimedes number. In the regime of $Re \ll 10^{-3}$ and $Pe \gg 10^3$, the dependance on the Reynolds and Peclet number can be neglected (Chang and Powell, 2002; Probstein et al., 1994; Shapiro and Probstein, 1992). The limits for this regime are determined by the Schmidt number, $Sc = \frac{Pe}{Re}$. As shown in Figure 1.1, the suspension is expected to be Newtonian

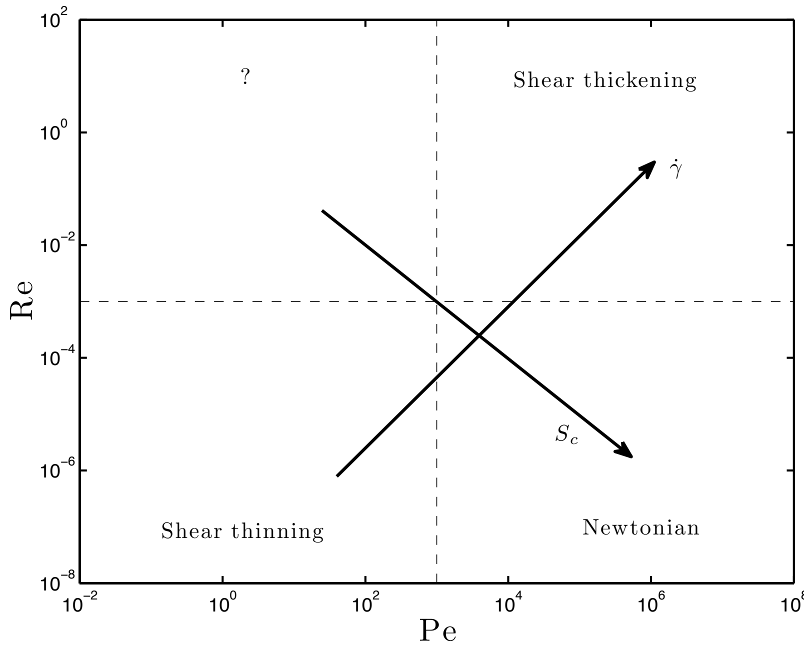

Figure 1.1: Flow pattern for suspension rheology, based solely on a dimensional analysis. Diagram from Stickel and Powell (2005).

for a certain range of shear rates and diminishing Schmidt number (Stickel and Powell, 2005). Under these conditions the relative viscosity of the suspension depends solely on the volume fraction,

$$\frac{\mu'}{\mu_f} = f(\phi).$$

Therefore, at a constant particle concentration, the suspension is expected to exhibit a constant relative viscosity and hence a Newtonian behavior. However, the effective viscosity of the suspension cannot be a function of the volume fraction alone. From the mixture theory viewpoint, the effective properties of the suspension depends on all the details of the microstructure and the particle concentration is not the only microstructure measure. Parameters like particle size distribution, temperature, porosity, etc. would determine the effective viscosity (Pabst, 2004). The extent of inertial effects is dictated by the Reynolds number. Stickel and Powell (2005) suggests that the shear thickening behavior observed in the review of Barnes (1989) and Hoffmann (1972) is due to such effects and considers that for $Re > 10^{-3}$, the inertial effects become important. The reason why shear thickening is linked to inertial effects is because when inertial effects are present, the interactions between particles increase and this changes the mechanism in which the momentum is transferred. The change in momentum due to collisions and the frequency of those collisions is proportional to the relative velocity of the particles, giving rise to stresses that depend on the square of the shear rate. However, the regime where this occurs is still not well established because it is difficult, as shown in the next section, to make direct comparisons between experiments.

In the following section, a description of the previous studies concerning suspensions with non-Brownian particles and Reynolds number higher than 1×10^{-3} is presented. The range of experimental parameters for the current study and those earlier studies are summarized in table 1.1.

Previous experiments [1]

As summarized by Koos et al. (2012), one of the earliest and most extensive experiments dealing with flows with Reynolds number greater than one was performed by Bagnold (1954). He used a small Couette flow rheometer with the inner cylinder fixed and the outer cylinder rotating. The top and bottom end caps were also rotating. Using this device he measured the shear and normal stresses for suspensions with a density ratio between the liquid and solid phase equal to one, over a range of shear rates and particle concentrations. He identified two flow regimes: (i) macro-viscous, where the suspension behaves like a Newtonian fluid and is considered non-collisional; and (ii) grain-inertia, where the interstitial fluid plays a minor role and the main contributions to the stresses are attributed to inter-particle friction and collisions.

The macro-viscous regime was found at low shear rates and the tangential stresses were linearly proportional to the dynamic viscosity of the liquid, the shear rate, and a function of solid fraction:

$$\tau \approx \mu \dot{\gamma} f_1(\phi).$$

The grain-inertia regime occurred at larger shear rates, similar to the results found for highly concentrated non-inertial suspensions, the stresses were independent of fluid viscosity and depended on the square of the shear rate, the square of the particle diameter, the particle density, and displayed a stronger dependence on the solid fraction:

$$\tau \approx {\rho_p}^2 d^2 \dot{\gamma}^2 f_2(\phi).$$

Bagnold distinguished the two regimes by defining a parameter, N, from the ratio of the scaling of the stresses,

$$N = \frac{\rho_p d^2 \dot{\gamma}^2 f_1(\phi)}{(\mu \dot{\gamma} f_2(\phi))} = \rho_p d^2 \dot{\gamma} g(\phi)/\mu,$$

where $g(\phi)$ is a function that increases with solid fraction. The parameter, N, has been referred to as the Bagnold number, and according to Bagnold $N \approx 450$ marked the transition from the viscous regime to the inertial regime. Zeininger and Brennen (1985) found a similar transitional Bagnold number for their hopper flows experiments. However, it is important to note that Bagnold's experiments involved neutrally buoyant particles so that the fluid and solid densities matched, $\rho_f =$

[1] Some of the material that is summarized in this section is taken from a paper by Koos, Linares-Guerrero, Hunt, and Brennen (2012)

ρ_p. In addition, Bagnold used only 1mm deformable wax beads. Hence, in these experiments only the shear rate and the fluid viscosities were varied; the density and the particle size were fixed.

Savage and McKeown (1983) tried to replicate Bagnold's experiments using a concentric cylinder rheometer. Their experimental setup differed from the one used by Bagnold. The flow was sheared by the rotation of the inner cylinder instead of the outer cylinder. Using a neutrally-buoyant liquid-solid suspension, they varied the particle size from 1 to 2 mm diameter and the roughness of the driving surfaces. They found higher shear stresses than Bagnold and although they found that shear stress varied with the square of the shear rate, they did not find a dependence on the square of the particle diameter. They concluded that the shear cells as well as the particles were sufficiently different from those used by Bagnold that direct comparisons cannot be made, even though the apparatus was just 3 cm higher and had a shear gap 6 millimeters wider than the one used by Bagnold. As it is discussed later in Section 1.3, rotating the inner cylinder is known to destabilize secondary flows (Mullin and Benjamin, 1980; Andereck et al., 1986; Conway et al., 2004). Hence, the work of Savage and McKeon suggested that the flow was governed not only by the length scale of the particles but also by the scale of the experimental apparatus.

One of the challenges that is encountered with inertial suspensions is that the experimental design may influence the measurements of the stresses. For example, in a flow with rotation, the radial inertia can induce a radial velocity within the flow (Taylor, 1936a,b). At low Reynolds number, the viscosity can suppress this radial motion, but as the Reynolds number increases, secondary flows, often referred to as Taylor vortices, may change the character of the flow field from a simple shear flow; this change in flow character can affect the measurement of the stresses. Hunt et al. (2002) made a detailed analysis of Bagnold's work and demonstrated that his experimental results were marred by the presence of secondary flows that developed at the boundaries of the rheometric device. By accounting for the contribution of the vortices to the shear stress, Hunt *et al.* demonstrated that Bagnold's linear to quadratic transition could be explained by assuming a laminar Newtonian flow with an effective viscosity that depended on the solid concentration. The presence of shear thickening can often be attributed to the improper design of the experiment, that is, machine artifacts mistaken for shear thickening. These phenomena could explain the results which otherwise show no relationship to other data in the literature, and does not dismiss the shear thickening behavior of concentrated suspensions.

Other experiments involving particles that are unaffected by Brownian motion have been done later are the ones done by Hanes and Inman (1985). They used glass spheres and water as the interstitial fluid in their rheological measurements ($\rho_p/\rho = 2.48$). They used an annular configuration where the sides and bottom rotated. The top did not rotate and was allowed to displace upwards in response to the normal stress generated by the mixture. They examined a narrow range of volume fractions between 0.49 and 0.58. The experiments used glass beads of two sizes in both water and air.

The particles used were denser than the surrounding fluid. The shearing surfaces were roughened by cementing one to two grain layers of the particles. According to the authors, the rotational speeds and applied normal stresses used in these experiments were selected to ensure that the centrifugal stress was always much less than the normal stress. The secondary flows were small compared with the primary flow and did not affect the stress measurements (Hanes and Inman, 1985). The shear stress was measured on the top wall. Their flow curves show slopes between 1 and 2, suggesting that the flow studied was in a transition regime. They observed the formation of a layer of static particles above which the granular material deformed rapidly. The presence of this layer suggests that the nature of the boundaries can have a significant effect upon the dynamics of the entire flow. In all of their experiments, the stresses were found to be weakly dependent on the volume concentration up to approximately 0.5, and strongly dependent above this concentration.

Similar to the work done by Acrivos et al. (1994), but with much larger particles ($d = 3.175$ mm vs $d = 0.14$ mm used by Acrivos *et al.*), Prasad and Kytömaa (1995) studied the transition between the quasi-static and the viscous regimes of dense particle suspension. They measured the effective viscosity of acrylic particles in an aqueous glycerine mixture ($\rho_p/\rho_f = 1.12$). In this study, only high solid fractions ($0.493 \leq \phi \leq 0.561$) were considered. They used an annular gap where the bottom was allowed to rotate and the top and sides remained fix. The stress measurements were made on the top surface. The upper and bottom walls were rough with a roughness scale of the order of the particle diameter. Their results show no evidence of secondary flows present ($9.3 \leq Re_b \leq 328$). Their device allowed them to conduct two kinds of experiments: constant normal stress and constant volume fraction. The first kind revealed that the maximum packing fraction (ϕ_m), defined as that solid fraction at which the mixture cannot be sheared, is not constant and increases with the normal stress, reaching a maximum with the shear rate. This observation was also done by Rutgers (1962) and later by Tsai and Gollub (2004). For the constant volume fraction experiments they found a shear thinning behavior for volume fractions above 0.543, and a Newtonian behavior for the set of runs below that value. Finally, they studied the influence of fluid viscosity by conducting experiments with a fluid viscosity one order of magnitude lower than that used in the earliest experiments. They did not find differences in their results, which is somewhat counterintuitive since the overall viscosity of the suspension is almost a direct function of the continuous phase viscosity.

In the recent years, a number of studies have attempted to unify the rheology of suspensions with the rheology of dry granular flows. However, such efforts have been limited to dense suspensions, where the volume fraction is higher than 50 %. Therefore, the link found between the two rheologies deals with the transition from friction dominated flow at low shear rates to viscous dominated at higher shear rates, rather than the transition from viscous dominated to inertia dominated flows. A summary of the studies regarding dense non-Brownian suspensions with moderate Reynolds number is presented below.

In a suspension with perfectly matched densities and volume fractions close to the random close packing, a presence of yield stress is expected to occur provided that normal stress is applied (Coussot and Ancey, 1999; Prasad and Kytömaa, 1995; Huang et al., 2005). However, the presence of yield stress has been observed in volume fractions looser than the random close packing. Fall et al. (2009) showed that the presence of yield stress in volume fractions lower than the random close packing is due to a density mismatch between the fluid and the solid particles. By carefully matching the two phase densities, rheological measurements on non-Brownian suspensions were performed using 40 μm in diameter polystyrene particles suspended in a NaI solution. They studied the effect of a slight density difference ($\Delta\rho = 0.15 g/cm^3$) and found that when the particles are perfectly buoyant, there is no presence of yield stress until volume fractions of 62%. Using magnetic resonance imaging in a wide-gap Couette geometry, they observed no presence of shear banding in suspensions with matched density, but for $\Delta\rho = 0.15 g/cm^3$, the flow is not homogeneous at low shear rates. As the shear rate increases the suspension becomes more homogeneous and the presence of shear bands disappear. This study suggests that the reason for the presence of yield stresses observed in the previous works at lower volume fractions is due to a slight density mismatch. Sedimentation or creaming may lead to the formation of more concentrated zones in which the particles are packed enough that a yield stress emerges. The presence of shear banding has also the same origin, where the normal stresses generated by the flow at low shear rates can no longer balance the gravity force. At higher shear rates, the shear induced resuspension of the particles prevent shear banding from occurring, making the flow to be homogeneous. In the current work, the effect of density mismatch is also studied where the presence of yield stress is observed, indicating that there is a slight difference in density for the matched density experiments.

Dijksmann et al. (2010) studied the rheology of non-Brownian suspensions with settling particles using a "split-bottom" geometry. Their experimental setup consisted on a square box with a rotating disk at the bottom. The radius of the disk is 4.5 cm and the width of the box is 15 cm. The suspension has a free surface the top and the height of the suspension is varied. They used acrylic particles with a diameter of 4.6 mm in an aqueous mixture of Triton X-100 and ZnCl2. The density ratio is $\rho_p/\rho = 1.1$. Based on the hypothesis that for vanishingly flow speeds, hydrodynamics effects are expected to be negligible and the behavior of the suspensions becomes similar to the behavior of dry granular materials. Dijksmann et al. (2010) derived a constitutive equation for their suspension using the modified inertial number approach proposed by Cassar et al. (2005),

$$\tau = A_o P + A_1 \frac{\mu\dot{\gamma}}{\alpha},$$

where P and α are pressure and porosity; and A_o and A_1 are empirical friction functions. Their constitutive model is reminiscent of the rheology of a Bingham fluid. Their results for suspensions sheared at low rotational speeds (driving rotating disk rates goes from $8.3 \times 10^{-5} \leq \dot{\gamma} \leq 8.2 \times 10^{-2}$ rps) compare favorably with the predicted flow field for slow dry flows, suggesting that the effect of interstitial fluid is negligible and that the mechanism of the suspension flow is dictated by friction. At higher shear rates, the flow behavior of the suspension is Newtonian and it compared favorably with the predicted flow field determined by a finite element software package. The stresses become rate dependent for driving rates of approximately 0.01 rps, and increase linearly with increasing shear rates. The torques measured for the suspensions in the Newtonian regime were compared with the torque for just the suspending liquid, and it was found that the effective viscosity of their suspension is only three to five times higher than the viscosity of the suspending liquid. Such values are far below what Krueger-Dougherty formula would predict. However, due to the non-confinement of their flow, the effective volume fraction of their suspension is not fixed, which complicates the analysis. The effect of interstitial fluid viscosity was also studied by using a different suspension with 2 mm glass beads and glycerol as the suspending liquid. The suspending liquid viscosity was varied by more than a decade by increasing the temperature from 4 to 37 $°C$. Their results show that once the measured torque is scaled with the change in viscosity, the data collapses into one single curve, where the flow exhibits a Bingham fluid-like behavior for low shear rates and a Newtonian behavior for higher shear rates. Decreasing the viscosity would also decrease the Reynolds number, and considering the highest driving rates tested, the range of Reynolds number goes from 1.26 to 26 for this set of experiments.

Boyer et al. (2011) made an analysis similar to Dijksmann et al. (2010) where the modified inertial number is used to unify the rheology of dense suspensions and dry granular flows. Using a pressure-imposed shear cell, measurements of the shear and normal stress were performed. The top boundary of their apparatus was free to displace in the vertical direction; thus the volume fraction tested varied accordingly to the normal pressure applied. At an initial stage, the volume fraction was chosen to be 56.5 % for most of their experiments. Two types of particles were used: (i) polymethyl methacrylate (PMMA) (d=1.1±0.05 mm) in triton X-100/water/zinc solution, and (ii) polystyrene (d=0.58 ±0.01 mm) in polyethylene glycol-ran-propylene glycol moonlitylether. The density ratio for both set of experiments was equal to one and settling effects were neglected together with the presence of particle migration. Using the modified inertial number (I_v) proposed by Cassar et al. (2005),

$$I_v = \frac{\eta(I_v)\dot{\gamma}}{P^p},$$

where P^p is the particle pressure, Boyer et al. (2011) proposed a constitutive model for the suspensions where the shear stress is proportional to the particle pressure. The coefficient of proportionality is given by the effective friction (η) that depends solely on the modified inertial number and can be inferred by the ratio of shear and normal stresses ($\eta = \tau/P^p$). For the two particles tested, the coefficient of friction for different particle pressures, liquid viscosity, and shear rates collapses into one single curve when plotted against I_v. The volume fraction can be calculated from the displacement of the top plate and it was found to be a decreasing function of I_v. Boyer et al. (2011) showed that this apparent frictional behavior of dense suspensions can be reconciled with the classical view that considers an effective viscosity if the suspensions is sheared at constant volume fraction, in which case, the shear and normal stresses depend on the viscosity of the fluid and the shear rate,

$$\tau = \mu_s(\phi)\mu\dot{\gamma} \text{ and } P^p = \mu_n(\phi)\mu\dot{\gamma},$$

where $\mu_s(\phi)$ and $\mu_n(\phi)$ are the dimensionless effective shear and normal viscosities. Equating the proposed constitutive equation with the classical view leads to the following relation:

$$\mu_s(\phi) = \frac{\eta[I_v(\phi)]}{I_v(\phi)} \text{ and } \mu_n(\phi) = \frac{1}{I_v(\phi)},$$

and therefore the rheology of the suspension is dictated by an effective viscosity that depends solely on the volume fraction and the effective friction. Their experimental results agrees with the correlation of Krieger and Dougherty for the limited range of volume fractions tested.

Trulsson et al. (2012) studied the transition from viscous to inertial regime in dense suspensions via numerical simulations, where it was found that the transition from Newtonian to shear thickening behavior is dictated by the ratio of the inertial number I defined as

$$I = \sqrt{\frac{\rho\dot{\gamma}^2 d^2}{P^p}},$$

and the modified inertial number I_v used by Boyer et al. (2011). In their simulations the lubrication interactions between the particles are considered and it was found that the transition from viscous to inertial regime is unaffected by it. The shear stress was found to be a sum of viscous forces and the forces due to particle interactions that depend quadratically with the shear rate, as proposed by Bagnold (1954),

$$\tau = f(\phi)(\mu\dot{\gamma} + k\rho d^2\dot{\gamma}^2)$$

where k is a constant of order 1 that encodes the details of dissipative mechanisms. While the work of Boyer et al. (2011) focused on the quasi-static viscous regime, the simulations of Trulsson et al. (2012) considered higher shear rates where the inertia becomes important. Trulsson et al. (2012) explained that the reason why the data of Boyer et al. (2011) collapses with just the modified inertial number is because they studied a regime with vanishingly shear rates.The quadratic dependance on the shear rate found by the numerical simulations of Trulsson et al. (2012) is limited to high volume fractions that are close to the jamming transition. In such case, the effective viscosity of the

Experiment	**d(mm)**	$\dot{\gamma}$ (s^{-1})	**Re**	**St**	ρ_p/ρ	**Flow behavior**
Bagnold (1954)[a] 50 % paraffin wax and lead stearate	1.32	7.34 - 52	14.94 - 89.62	1.66 - 9.95	1	Newtonian-Shear thickening
Savage *et al. (1983)*						
polystyrene	0.97	7.86 - 59.78	7.12 - 54.09	0.79 - 6.01	1	Newtonian-Shear thickening
	1.24	5.018 - 71.15	7.42 - 105.22	0.82 - 11.69		
	1.78	8.04 - 65.57	24.50 - 199.79	2.722 - 22.20		
Hanes *et al. (1985)*						
glass beads	1.1	35.60 - 263.00	43 - 317	11.88 - 87.65	2.48	Newtonian-Shear thickening
	1.85	34.4 - 127	118.00 - 434.00	36.37 - 134.25	2.78	Newtonian-Shear thickening
Acrivos *et al (1994)*						
PMMA [a]	0.1375	0.45 - 5.374	8×10^{-4} - 3×10^{-2}	8×10^{-5} - 3×10^{-3}	1	Shear thinning
acrylic	0.0905	0.135 - 44.13	3×10^{-6} - $9.\times10^{-4}$	3×10^{-7} - 1×10^{-4}	0.95	Shear thinning
Prasad *et al. (1995)*						
glass beads	2	1 - 100	1.37-13.7	0.32-3.2	2.09	Shear thinning - Newtonian
acrylic	3.175	0.50 - 16.00	0.09 - 2.89	0.01 - 0.36	1.12	Shear thinning - Newtonian
Fall *et al. (2009)*						
polystyrene	0.04	$1\times10^{-4}-10$	$4\times10^{-6}-4\times10^{-3}$	$5\times10^{-7}-5\times10^{-4}$	0.85 and 1	Viscoplastic
Dijksman *et al. (2010)*						
acrylic	4.6	$2\times10^{-3}-10.4$	$4\times10^{-4}-0.72$	$5\times10^{-5}-0.09$	1.1	Viscoplastic-Newtonian
glass	2	$5\times10^{-3}-31.4$	$6\times10^{-5}-2.4$	$1\times10^{-5}-0.53$	$1.88-1.92$	Viscoplastic-Newtonian
Boyer *et al. (2011)*						
polystyrene	0.58	0.1-2.5	$1\times10^{-5}-4\times10^{-4}$	$2\times10^{-6}-5\times10^{-5}$	1	Viscoplastic-Newtonian
PMMA	1	0.1-2.5	$4\times10^{-5}-1\times10^{-3}$	$4\times10^{-6}-1\times10^{-4}$	1	Viscoplastic-Newtonian
Koos *et al. (2012)*						
polystyrene	3.34	2.19 - 74.87	18.05 - 782.04	2.04 - 89.46	1 and 1.009	Newtonian
nylon	6.36	7.77 - 70.24	50.36 - 718.31	5.65 - 81.21	1	Newtonian
SAN [b]	3.22	4.72 - 83.04	23.21 - 539.08	2.58 - 60.19	1	Newtonian

Table 1.1: Previous experiments properties.

[a] Macro-viscous regime
[a] polymethyl methacrylate
[b] styrene acrylonitrile

suspension diverges leading to vanishingly Reynolds numbers. Therefore, the transition from the viscous to the inertial regime at high volume fractions is of a different nature that the transition formulated by Bagnold (1954), where particle collisions control the regime.

Kulkarni and Morris (2008), Yeo and Maxey (2013), and Picano et al. (2013) studied the rheology of inertial suspensions for a wider range of volume fractions by means of numerical simulations. In these three studies the Reynolds number was larger than 10^{-3} and both the inertia of the particles and the fluid are considered. The simulations considered a density ratio equal to one. The common result between these simulations is the increase of effective relative viscosity with Reynolds number. Figure 1.2 shows the results from these numerical works.

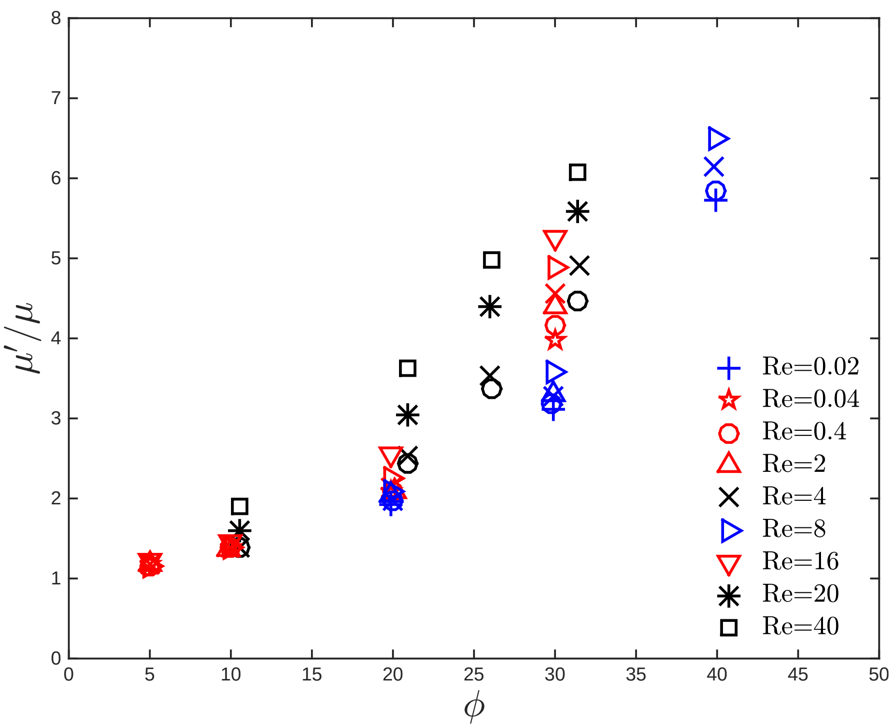

Figure 1.2: Predicted effective relative viscosity for inertial suspensions from numerical studies. Symbols denote the Reynolds numbers according to figure legend. Red symbols are from Kulkarni and Morris (2008), blue symbols are from Yeo and Maxey (2013), and black symbols are from Picano et al. (2013).

The previous work of Koos et al. (2012) showed a different result. In the range of Reynolds number tested (20 to 800), the effective viscosity of the suspensions with neutrally buoyant particles showed no dependance on the Reynolds number. The reason for this difference in results is still not clear but it is believed that the transition from macro viscous to inertial regime is dictated by the Stokes number and that for the range of Stokes numbers tested the particle interactions are damped by viscous dissipation of the interstitial fluid. A detailed description of this study is presented in Chapter 3.

The present work is a continuation of the work done by Koos et al. (2012). Koos performed rheological measurements of liquid solid mixtures with relatively large particles (ranging from 3.34 to 6.36 mm) with a density ratio of one. The range of Reynolds and Stokes numbers tested were between $20 \leq Re \leq 800$ and $3 \leq St \leq 90$ respectively. The effective viscosity for all the mixtures tested showed no dependance with the Reynolds and equivalently with Stokes numbers. A further detail of the work of Koos is presented in Chapter 3.

Table 1.1 shows the regimes for this kind of suspensions and the flow behavior observed in previous experiments. Based on the phase diagram proposed by Stickel and Powell (2005), all the mixtures were expected to exhibit a shear thickening behavior. This suggests that this phase diagram needs to be re-scaled. Based on Table 1.1, a suspension may be expected to exhibit a Newtonian behavior for greater ranges of shear rates as particle size and fluid viscosity increase.

The diagram shown in Figure 1.3 summarizes the Reynolds and volume fractions regions studied in the previous experimental studies along with the range covered in the current work.

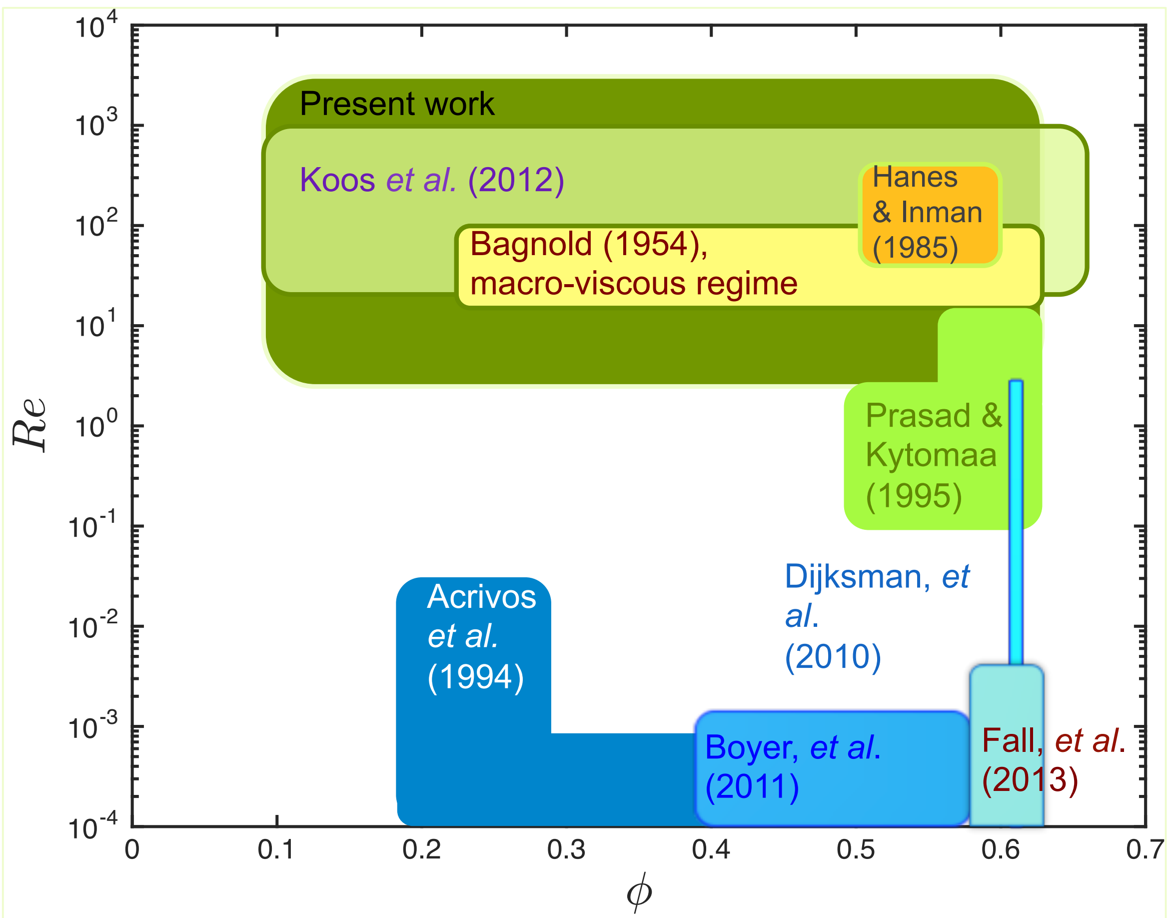

Figure 1.3: Diagram of previous and current experimental work done in inertial suspensions in terms of Re. The properties of the previous experiments can be found in table 1.1

Particle interactions

The Stokes number is a dimensionless number that gives a measure of the ratio of available momentum in the solid phase that sustains the particle motion through the liquid to the liquid viscous forces (Yang and Hunt, 2006). In several studies (MacLaughlin, 1968; Davis et al., 1986; Joseph et al., 2001; Joseph and Hunt, 2004; Yang and Hunt, 2006), the Stokes number has been used to characterize the collision of particles immersed in a viscous fluid. The energy loss during the collision is described by the effective coefficient of restitution (e). It has been found that with diminishing Stokes number, a monotonic decrease in e is observed (Joseph et al., 2001; Joseph and Hunt, 2004; Yang and Hunt, 2006). This dependence is the same for normal and oblique collisions between pairs of identical and dissimilar spheres, as well as for sphere-wall collisions. For St>2000, the effect of the fluid becomes negligible, resulting in a nearly unity restitution coefficient that approximates a dry impact. However, with increasing liquid viscosity or decreasing particle inertia, the sphere can no longer sustain its motion through the liquid and a critical particle Stokes number, St ≈ 10, exists below which no rebound occurred. These findings suggest that for dense suspensions there will be a critical Stokes number below which the collisions of the particles will be damped by the liquid, and above it there will be a decrease of energy lost during the particle collisions.

The Stokes number tested in the current experiments vary from 2.5 to 195 and it is expected that the particle collisions in these regime become important.

The diagram shown in Figure 1.4 summarizes the Stokes and volume fractions regions studied in the previous and in the present experimental work.

Particle settling

When the particles are denser than the suspending liquid, the particles can settle depending on the shearing conditions and the density ratio. The mechanical properties of liquid-solid flows can be strongly affected in the presence of settling since the mixture is no longer homogeneous. When sheared, the particles can re-suspend. The resuspension of particles at vanishingly Reynolds number was first observed by Gadala-Maria and Acrivos (1980). This phenomena called *viscous resuspension* has been modeled by balancing gravitational and shear induced particle migration by Leighton and Acrivos (1986) and it was successfully employed by Acrivos et al. (1993). In the model of Leighton and Acrivos (1986), the particle terminal velocity (v_{ter}) was given by Stokes flow:

$$v_{ter} = \frac{2}{9}\frac{a^2 g(\rho_p - \rho)}{\mu}$$

where a is the particle radius and g is the gravitational acceleration.

The particles can be fluidized with increasing shear rate. King (2001) categorizes the liquid-solid

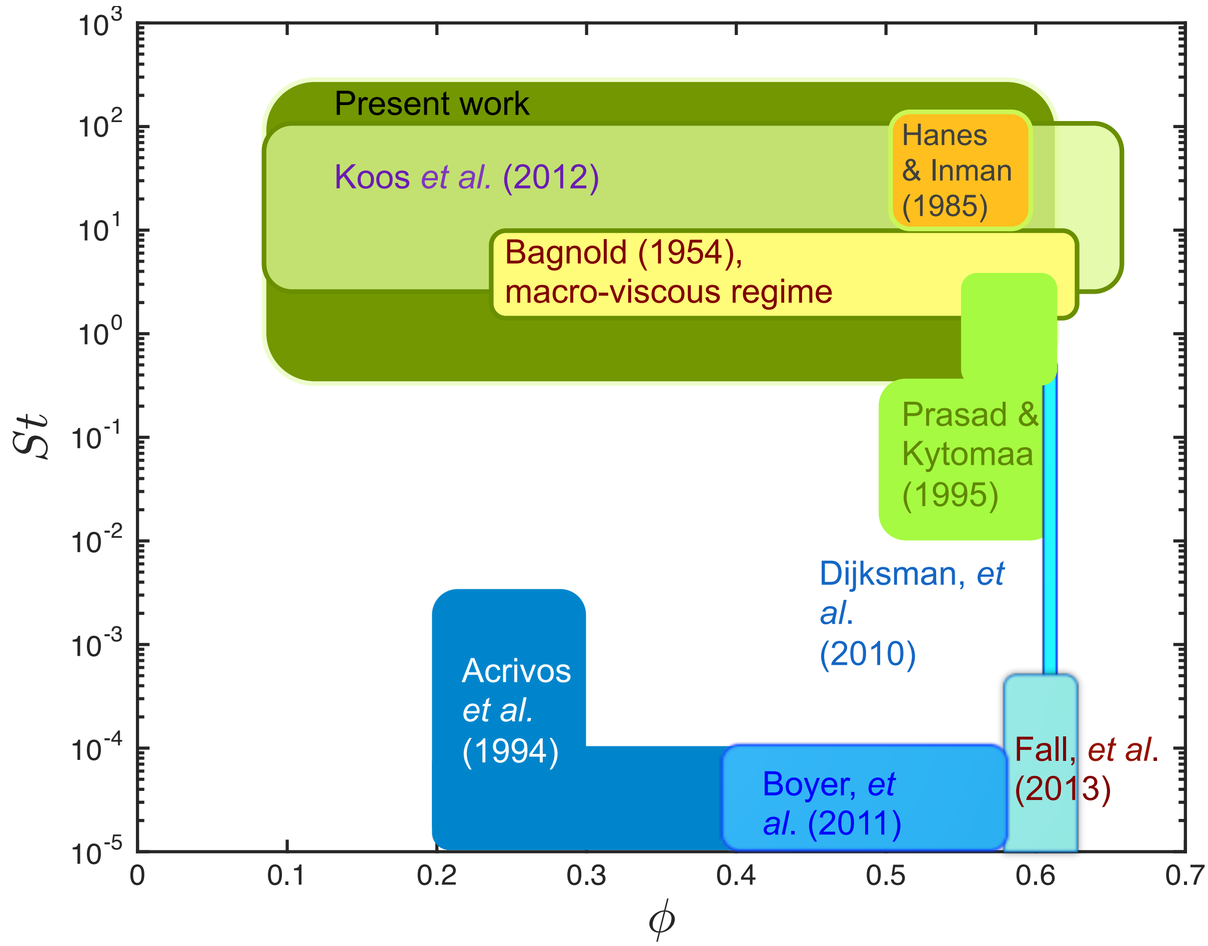

Figure 1.4: Diagram of previous and current experimental work done in inertial suspensions in terms of St. The properties of the previous experiments can be found in table 1.1

flows based on variation of Archimedes number and Reynolds number, where the suspension would be homogenous for low Ar and moderate Re, and become heterogeneous with increasing Ar. As the ρ_p/ρ increases, the required Reynolds number to fluidize the particles also increases (Bi and Fan, 1992; King, 2001). The effective viscosity of the suspension depends strongly on the volume fraction which can vary when the particles are denser than the liquid. To account for such variations, in the present experiments visualizations of the flow are made allowing to infer the local volume fraction and correlate it to the measured effective viscosity.

1.3 Secondary flows

In a rotating flow, instabilities due to the deviation from the azimuthal direction of the flow occur. This deviation is primarily due to centripetal forces generated in a rotating fluid. Taylor (1936a) observed that in a coaxial cylindrical Coquette flow, counter rotating laminar tori develop. When the flow was driven by the rotation of the inner cylinder, the development of highly ordered patterns formed after the inner cylinder rotation rate exceeded a critical value. When the flow was driven by the rotation of the outer cylinder and the inner cylinder remained fixed, such transitions occurred more gradually. Under this configuration, the formation of a recirculating flow at the top and bottom boundaries occurred. Taylor (1936a,b) observed that under the rotation of the outer cylinder the circulatory motion is stable to infinitesimal disturbances and also that there is an intermediate range of Reynolds number in which the flow is in transition and stable. Instabilities of the flow occur at much lower Reynolds numbers when the inner cylinder drives the flow due to centrifugal forces. Conway et al. (2004) observed the formation of Taylor like vortices in a dry granular flow at slightly lower Reynolds number, proving that such instabilities are also likely to occur in granular materials.

The presence of these flows increases the shear stress on the cylinder walls. Coles (1965) showed that the measured torque under the presence of secondary flows can increase in a nonlinear manner. Therefore, care should be taken to avoid the effect of the presence of these secondary flows when acquiring rheological data. In the current work, the rheometer used was specifically designed to delay the presence of such effects. However, how the presence of particles influence the strengthening or weakening of these flows is difficult to estimate. Matas et al. (2003) summarized the effects of the presence of particles in a horizontal pipe flow. The transition from laminar to turbulent depends on the ratio between the particle and pipe diameter. When this ratio increases, the critical Reynolds number decreases for volume fractions less than 10, and it increases as the volume fraction increases and the ratio of particle and pipe diameter increases. Gore and Crowe (1991) observed that turbulence was strengthened by small particles and attenuated by large ones.

Effect of rough boundaries

To avoid slip at the wall (a condition where the solid phase of the mixture can roll and slide at the wall or move away from it), the current experiments were performed using rough walls. The presence of rough boundaries can impact the stability of the fluid. Cadot et al. (1997) performed experiments on Couette-Taylor flow with smooth and rough walls. In these experiments the flow was driven by either counter-rotating the cylinders or by the rotation of the inner one. The presence of roughness at the wall did not change the transition but it did increase the measured torques after the threshold was reached. Similar to what was found by Cadot et al. (1997), van den Berg et al. (2003) and Lee et al. (2009) examined the differences in transition for walls with different roughness and found that the critical Reynolds number is the same regardless of the presence or absence of roughness.

All these studies considered the rotation of the inner cylinder, where the transition occurs abruptly and the flow is more unstable. It is possible that under the rotation of the outer cylinder the range of Reynolds number at which the flow is in transition but stable can be affected by the presence of roughness.

1.4 Thesis outline

The primary objective of this thesis is to investigate the inertial effects in liquid-solid flows. The current work is a continuation of the work done by Koos (2009), and emphasis has been focused on extending the Stokes and equivalently the Reynolds number regime studied previously via experiments. Rheological measurements using a concentric cylinder apparatus equipped with rough walls were performed for particles with the same or higher density than the suspending liquid.

The description of the rheometer and the methods for measuring the rheological properties of the liquid-solid flow is presented in Chapter 2. Measurements of the torque for pure fluid (no particles) is also presented where the presence of hydrodynamics instabilities is discussed and analyzed. The results from the previous experimental work done by Koos et al. (2012) is reported in Chapter 3. In Chapter 4 the torque measurements for particulate flows with density ratio (ρ_p/ρ) equal to 1 and 1.05 are described. Chapter 5 presents the results for flows with $\rho_p/\rho = 1.05$ over a porous medium.

For the experiments with settling particles, a characterization of the particle resuspension is performed and presented in Chapter 6. The measurements of the settling particles' expansion are used to predict the effective volume fraction for the experiments with $\rho_p/\rho = 1.05$. Chapter 7 presents a discussion and analysis of the obtained results. A comparison between the results with different density ratio is presented and the effect of hydrodynamics instabilities is analyzed.

Chapter 8 summarizes the results obtained from this investigation. Topics for future work are discussed as well.