Mixing behavior of a model cellulosic biomass slurry during settling and resuspension

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Abstract

Thorough mixing during biochemical deconstruction of biomass is crucial for achieving maximum process yields and economic success. However, due to the complex morphology and surface chemistry of biomass particles, biomass mixing is challenging and currently, it is not well understood. This study investigates the bulk rheology of negatively buoyant, non-Brownian α -cellulose particles during settling and resuspension. The torque signal of a vane mixer across two distinct experimental setups (vane-in-cup and vane-in-beaker) was used to understand how mixing conditions affect the distribution of biomass particles. During experimentation, a bifurcated torque response as a function of vane speed was observed, indicating that the slurry transitions from a “settling-dominant” regime to a “suspension-dominant” regime. The torque response of well-characterized fluids (i.e., DI water) were then used to empirically identify when sufficient mixing turbulence was established in each experimental setup. The predicted critical mixing speeds were in agreement with measured values, suggesting that secondary flows are required in order to keep the cellulose particles fully suspended. In addition, a simple scaling relationship was developed to model the entire torque signal of the slurry throughout settling and resuspension. Qualitative and semi-quantitative agreement between the model and experimental results was observed.

Keywords: Biomass, Cellulose, Resuspension, Settling, Mixing, Rheology

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1. Introduction

The conversion of biomass, specifically cellulosic biomass, into liquid transportation fuels (i.e., biofuels) has the potential to provide renewable alternatives to petroleum-based products, such as gasoline, diesel, and jet fuel. The production of biomass-derived fuels can increase energy security, reduce CO₂ emissions, and fit seamlessly into existing liquid transportation fuel infrastructure [1, 2]. However, current biochemical deconstruction technologies and processing strategies need further improvement in order to make cellulosic biofuels cost competitive with petroleum-based fuels.

The deconstruction of cellulosic biomass into sugar-rich intermediates involves multiple slurry-phase process streams [3–6]. In order to reduce processing costs, the concentration of biomass species (insoluble and soluble) within these slurry streams should remain as high as possible [7–9]. High solids concentrations can reduce capital and operating costs by decreasing vessel volumes and lowering heat usage. Additionally, the amount of water that must be removed during energy-intense, back-end processing and product purification is greatly reduced [10]. During the early stages of biomass deconstruction at high solids concentrations, the slurries are thick and paste-like, making the transport of these fibrous suspensions very challenging. After hydrolysis and liquefaction, the slurries have lower concentrations of insoluble solids and exhibit settling behavior. Proper mixing during biomass deconstruction is vital to achieving maximum process yields and economic success. Inadequate mixing can lead to disproportionate distributions of heat, chemicals, and enzymes, resulting in product degradation and enzyme inhibition [11]. Therefore, a more complete understanding of the mixing and transport behaviors of biomass slurries is needed for improved mixing strategies and reactor design.

Further complicating their rheological behavior, biomass slurry particles are porous, anisotropic, deformable, and generally non-Brownian. Recent studies have focused on examining the viscosity [11] and yield stress of these materials [12–14], and the feasibility of using rheology modifiers to reduce yielding behavior [15, 16]. However, there has been a limited number of studies on the mixing and settling of insoluble biomass particles during mixing.

Dispersions of non-Brownian particles in Newtonian fluids (i.e., granular suspensions) display a wide array of rheological behaviors: yield stress [17–20], shear banding [17, 19, 21], shear thickening [22, 23], and normal stress dilation [24–26], to name a few. Most important to the work presented here is

the phenomenon known as resuspension; the process in which an initially settled layer of negatively buoyant particles are remobilized and convected into the surrounding bulk fluid. Early work by Leighton and Acrivos [27] investigated the viscous resuspension of glass spheres, where the downward flux of particles was balanced by an upward flux resulting from shear-induced diffusion. Since then, authors have investigated viscous resuspension of spherical particles, both experimentally and theoretically, for flow in pressure-driven tubes [28–30] and channels [31–33], cylindrical Couette devices [34, 35], rotating parallel plates [36–38], and inclined planes [39–41]. However, the precise conditions for which a settled layer of particles will resuspend or, in contrast, when particles in a suspension will begin to settle, are not fully understood, especially with non-spherical, deformable, and porous particles.

Recent experimental studies of non-density-matched suspensions alluded to the existence of two distinct flow regimes during resuspension. At low shear speeds, interparticle contact is high and the rheology is governed by the frictional interactions between particles (resembling granular solids). At high shear speeds, the particles are resuspended, lose contact, and display Newtonian flow behavior [17, 19, 20, 42, 43]. Work by Fall et al. [19, 20] has attempted to answer the question: what is the critical strain rate at which the transition between these two regimes occurs? Using a simple stress balance and dimensional analysis, Fall et al. [19, 20] suggest that the critical strain rate for the transition between granular and Newtonian behavior is dependent on a critical length scale. However, the question still remains, what is this “length scale?” Fall et al. first reported that the critical length was the particle radius [19] and then later discovered that the critical rotation rate to homogenize a suspension of non-Brownian particles is closely related to the filling height of the suspension [20].

In the work presented here, we investigate the global rheology of negatively buoyant, non-Brownian α -cellulose particles during settling and resuspension. The torque signals of a vane mixer from two unique experimental setups (vane-in-cup and vane-in-beaker) were used to further understand how mixing conditions affect the distribution of biomass particles. Our goal is to use these torque measurements and visualization at varying mixer speeds as a way to quantify and characterize particle resuspension, mixing effectiveness, and mixer power requirements. To our knowledge, this is the first study that investigates the resuspension of non-colloidal, non-spherical, deformable, and porous particles. Beyond this manuscript, we are interested in investigating the settling and resuspension behavior of biomass slurries using computa-

tional fluid dynamics (CFD). However, we first need a comprehensive set of experimental data in order to validate our ongoing modeling efforts. The results presented here serve as a foundation for future exploration into mixing and resuspension dynamics using extensive CFD modeling. In general, this work (as well as future CFD studies) will help better understand the torque requirements to achieve a well-mixed dispersion of biomass solids, in hopes of guiding engineers towards improved bioreactor design and enhanced biomass slurry processing.

2. Experimental Methods

2.1. Materials

For this study, α -cellulose obtained from Sigma-Aldrich Corporation (St. Louis, MO) was suspended in neat DI water at a concentration of 5%, by weight. The density of α -cellulose is $\sim 1500 \text{ kg/m}^3$ (as stated by the manufacturer), resulting in negatively buoyant particles when dispersed in DI water, which has a density of $\sim 1000 \text{ kg/m}^3$. The particle size distribution was measured using laser diffraction (Mastersizer 2000 Hydro, Malvern Instruments, Malvern, UK), where size is reported as a volume-equivalent spherical diameter. The α -cellulose particles have a broad size distribution, with spherical-equivalent diameters ranging from ~ 10 - $1000 \mu\text{m}$ (Figure 1a). The mean volume-equivalent spherical diameter measured with laser diffraction was $142 \mu\text{m}$, while the 10th, 50th and 90th percentiles of the cumulative particle size distribution were 22, 79, and $381 \mu\text{m}$, respectively. In addition, optical microscopy imaging (Leica DM 3000, Clemex Technologies Inc., Longueuil, Quebec, Canada) shows that the α -cellulose particles are heterogeneous in size and shape, and are rod-like with a wide range of aspect ratios (Figure 1b). The broad and bimodal-like size distribution in Figure 1a is most likely a direct result of the non-sphericity and heterogeneity of the α -cellulose particles (laser diffraction measurements can be impacted by particle orientation).

2.2. Rheometry

Torque measurements were conducted using an AR-G2 stress-controlled rheometer (TA Instruments, New Castle, DE). In order to avoid slip at the sample-tool interface, the four-blade vane rotor was used instead of the smooth-surface cylindrical bob. Figures 2a and b show the vane fixture, which is constructed of stainless steel. The vane blades are 38.25 mm in

Figure 1: (a) Particle size distribution of α -cellulose measured with laser diffraction. (b) Optical-microscopy image of α -cellulose particles.

111 height and 1.25 mm thick, with an overall tool diameter of 15 mm. In ad-
112 dition to the 15 mm vane, a larger vane (28 mm in diameter) of similar
113 construction was also used to help corroborate the empirical modeling ef-
114 forts presented in Sections 3.3 and 3.4. Unless otherwise noted, the data
115 presented are from the smaller, 15 mm vane rotor.

116 Rheological experiments were performed using two different setups: the
117 conventional vane-in-cup geometry (Figure 2c) and the custom vane-in-beaker
118 arrangement (Figure 2d). The vane-in-cup setup utilizes TA Instruments’
119 standard stainless steel cup (30 mm inner diameter), with temperature con-
120 trol of ± 0.1 °C provided by the heating jacket. The vane-in-cup geometry
121 was operated at 20 °C with the vane positioned 4 mm from the bottom of the
122 cup, which required 43 mL of sample for complete submersion of the vane
123 fixture.

124 In order to visualize the sample during shearing, the standard cup was
125 replaced with a 100 mL glass beaker (70 mm height and 47 mm diameter;
126 Fisher Scientific, Pittsburgh, PA) fastened to the rheometer Peltier plate
127 with adhesive putty (3M, St. Paul, MN). The vane-in-beaker setup required
128 a sample volume of 80 mL for full submersion of the vane. Again, the vane
129 was positioned 4 mm from the bottom of the beaker.

130 For both of the experimental setups, a stepped speed ramping procedure
131 was employed. The angular speed of the vane was increased (after start-up
132 from rest) from 1 to 45 rad/s, for which 14 speed steps were investigated. In
133 order to investigate any hysteresis effects, the angular speed was stepped back
134 down from 45 to 1 rad/s immediately following the increasing speed ramp.
135 Each speed was held constant for a minimum of 1 min and a maximum of 10
136 min, and data was gathered every 3 sec, allowing the mixing and settling of
137 particles to reach steady state. Longer shearing periods (up to 60 min) were
138 investigated, but no additional changes in mixing behavior were observed
139 (steady state was typically reached within 1 to 5 min of mixing at a given
140 speed). The system was considered to have achieved “steady state” once
141 the instrument recorded 5 consecutive torque measurements all within 5% of
142 one another. Rheometer data was averaged over the 15 sec data collection
143 window (5 points collected at 3 sec intervals), yielding a single torque value
144 for each input speed. As soon as steady state was reached, the instrument
145 moved sequentially to the next rotation rate in the predetermined window of
146 input speeds. Each experiment was repeated in triplicate (at minimum) to
147 measure reproducibility and quantify experimental variance.

148 The stepped angular speed ramping procedure was performed on fully

Figure 2: (a) Profile and (b) bottom view of the four-blade vane rotor (38.25 mm in height, 15 mm in diameter, and 1.25 mm thick). (c) Vane-in-cup and (d) vane-in-beaker experimental setups. The 5 wt% α -cellulose slurry is in the fully settled state in image (d).

Figure 3: Steady torque as a function of increasing (filled triangles) and decreasing (open triangles) vane rotation rate for an initially fully settled (16 hrs) 5 wt% α -cellulose slurry in the vane-in-cup setup. The torque response of the slurry is compared to the torque signal of the DI water control (filled diamonds).

149 settled 5 wt% α -cellulose dispersions. The fully settled samples were obtained
 150 by letting the particles settle out of suspension and equilibrate for 16 hrs
 151 before commencing vane rotation.

152 3. Results and Discussion

153 3.1. Vane-in-Cup Mixing

154 Figure 3 shows the mixing of a fully settled 5 wt% α -cellulose slurry
 155 in the vane-in-cup setup, which displays a bifurcated torque response as a
 156 function of mixer speed. Initially, the torque signal of the vane at 1 rad/s
 157 was relatively high ($\sim 70 \mu\text{N}\cdot\text{m}$) as the rotor slowly sliced through the settled
 158 particle bed at the bottom of the rheometer cup. As the vane speed was
 159 increased from 1 to 15 rad/s (Figure 3; filled triangles), the torque response
 160 decreased, reaching a minimum measured torque ($\sim 7 \mu\text{N}\cdot\text{m}$) at 15 rad/s.

161 When the vane rotation rate was further increased from 15 to 45 rad/s, a
 162 monotonic increase in the torque response was observed. The rise in torque
 163 signal from 15 to 45 rad/s mirrors the torque response of the DI water control
 164 (Figure 3; filled diamonds), indicating Newtonian behavior at rotation rates
 165 above 15 rad/s. Previous rheological studies of granular suspensions also

166 observed Newtonian flow behavior at elevated shear speeds when the solids
167 became resuspended [17, 19, 20, 42, 43]. Therefore, the inflection point in
168 the torque signal is believed to indicate a transition between the “settled
169 regime” and “suspended regime” (identified with arrows in Figure 3).

170 In order to assess any hysteresis effects, the vane speed was reduced from
171 45 to 1 rad/s (Figure 3; open triangles) immediately following the increasing
172 speed ramp. The torque signals from the increasing and decreasing speed
173 ramps were nearly identical, indicating little system memory. As described
174 above, prior to the increasing speed ramp, the α -cellulose slurry was left stag-
175 nate for 16 hrs allowing the particles to completely settle out of suspension,
176 forming a perceived dense bed at the bottom of the rheometer cup. However,
177 during the decreasing speed ramp, the particles settled out of suspension as
178 data was being collected (on the time scale of minutes, not hours) and the
179 settled bed was assumed to be somewhat “fluidized” and not densely com-
180 pacted. Yet, the flow curves from the increasing and decreasing speed ramps
181 in the vane-in-cup setup were not impacted by the settling history of the
182 solids bed.

183 3.2. Vane-in-Beaker Mixing

184 In order to visualize and help corroborate the above observations in mix-
185 ing behavior, the stainless-steel rheometer cup was replaced with a clear, 100
186 mL laboratory beaker. Due to the large spacing between the outer edge of
187 the vane and the inner wall of the beaker, the vane-in-beaker setup is not an
188 ideal rheometer geometry. However, it more-closely mimics a true bioreac-
189 tor system, where the flow field is not well defined and unmixed or “dead”
190 volumes commonly arise due to the large gap between the rotor and vessel
191 wall.

192 Using this vane-in-beaker setup and the same speed ramping protocol,
193 still images of the α -cellulose dispersion were captured *in situ* with rheologi-
194 cal measurement (Figures 4 and 5). In general, Figure 4 shows that a similar
195 diverged torque response was observed for the 5 wt% α -cellulose slurry us-
196 ing the vane-in-beaker setup as witnessed with the vane-in-cup setup (Figure
197 3). However, during the increasing speed ramp in the vane-in-beaker setup
198 (Figure 4; filled squares), the torque signal in the settled regime decreases
199 dramatically and the torque response of the α -cellulose slurry is equivalent
200 to that of the DI water control (filled circles) from 11 to 15 rad/s. The drop
201 in torque indicates that the α -cellulose particles are pushed away from the
202 vane during the increasing speed ramp and a localized environment depleted

Figure 4: Steady torque as a function of increasing (filled squares) and decreasing (open squares) vane rotation rate for an initially fully settled (16 hrs) 5 wt% α -cellulose slurry in the vane-in-beaker setup. The torque response of the slurry is compared to the torque signal of the DI water control (filled circles). The letters (a) through (h) correspond to the still images in Figure 5.

of particles arises near the vane interface. The depletion of particles near the tool wall is commonly observed during the rheological interrogation of two-phase dispersions [44] and recently, Ovarlez et al. [45] used magnetic resonance imaging to visualize the depletion of particles at the vane-sample interface (claiming that this is an unavoidable effect when studying the dynamic flow of large particle ($\sim 100 \mu\text{m}$) dispersions).

The resuspension and subsequent settling of particles near the beaker wall can be visualized in Figure 5, images (a) through (h). Initially at 1 rad/s (Figure 5a), all of the α -cellulose particles have settled into a compacted solids bed at the bottom of the beaker and a transparent liquid layer is evident above the solids. As the rotation rate of the vane is increased to 5 rad/s (Figure 5b), particles are convected into suspension, the height of the solids bed decreases, and the top liquid layer becomes slightly opaque. Next, as the speed is increased to 9 rad/s (Figure 5c), additional particles are resuspended and the liquid layer becomes even cloudier, obscuring the outline of the vane rotor. However, the height of the solids bed near the beaker wall increased and upon close inspection, two distinct layers of settled particles were observed (Figure 5c). The bottom layer consisted of the original settled

Figure 5: Still images of a 5 wt% α -cellulose slurry as a function of increasing vane speed (a-h). Images (a) through (h) correspond to the torque measurements displayed in Figure 4. The arrows in image (d) signify the two layers of settled solids.

particles, which was about equal height to the particle bed in Figure 5b. The top layer, on the other hand, consisted of freshly deposited particles (the two layers are identified with arrows in Figure 5d).

As the vane rotation rate was then increased to 11 rad/s, the deposited solids near the beaker wall reached a maximum height (Figure 5d) and were still comprised of two separate layers: layer one (the original settled bed height observed at 5 rad/s, Figure 5b) remained unchanged, while the second layer (consisting of freshly deposited particles) was enlarged. Simultaneously, the vane torque also dropped substantially at 11 rad/s and became equivalent to the torque signal of DI water (Figure 4). This drastic drop in torque coupled with the increase in freshly deposited solids suggest that a significant amount of particles were convected away from the vane and were then trapped in the low-shear regime near the beaker wall.

The particle-bed height eventually began to decrease as the vane speed was increased to 13 and 15 rad/s (Figure 5e and f). The original bottom layer remained intact, while the newly deposited top layer began to erode away. However, the torque signals at 13 and 15 rad/s (Figure 4) were still equivalent to that of DI water. As the vane speed was increased to 20 rad/s,

the torque signal of the α -cellulose suspension finally began to deviate from the DI water response (Figure 4) and the top layer of the solids bed had been completely eroded (Figure 5g). When the speed was increased again to 25 rad/s, the solids bed was nearly completely resuspended (Figure 5h) and the torque response of the 5 wt% α -cellulose slurry was significantly higher than the torque signal of DI water.

As the vane rotation rate was increased from 25 to 45 rad/s in the vane-in-beaker setup, the same monotonic increase in the torque signal was observed for the α -cellulose slurry as previously noted for the vane-in-cup system; signifying Newtonian-like behavior for the fully suspended granular dispersion at high vane speeds.

Immediately following the increasing speed ramp, the speed was then reduced from 45 to 1 rad/s and a significant hysteresis effect was observed (Figure 4; open squares). During the decreasing speed ramp, the torque signal remained larger than during the increasing ramp (mostly in the settled regime). Unlike the flow curves from the vane-in-cup system, it appears that the compaction or settling history of the solids bed impacted the torque response in the larger-gap vane-in-beaker setup. During the increasing ramp, only the particles on the interior of the solids bed (nearest the vane interface) were impacted by the vane's rotation, while the compacted solids near the beaker wall remained undisturbed until the vane's rotational speed reached ~ 25 rad/s (Figure 5h). Conversely, as the rotation rate was reduced from 45 to 1 rad/s and particles began to settle out of suspension, the particle bed remained fluidized and the vane's rotation influenced a larger fraction of the solids bed; leading to an increased torque signal during the decreasing speed ramp.

In order to further investigate the observed hysteresis effect in the vane-in-beaker system, the transient torque signals for the 11 rad/s step for both the increasing and decreasing speed ramps were examined (Figure 6). The 11 rad/s step was selected because this specific rotation rate showed the largest discrepancy in torque signal between the increasing and decreasing speed ramps in Figure 4. During the increasing speed ramp (Figure 6; filled squares), the torque signal is consistent at $1.6 \pm 0.1 \mu\text{N}\cdot\text{m}$ and remains unchanged over the 10 min time period. However, during the decreasing speed ramp (Figure 6; open squares) from 3 to 150 sec, the torque signal initially increases from 4 to 14 $\mu\text{N}\cdot\text{m}$. From 150 to ~ 540 sec, the torque response slowly decreases to $\sim 9 \mu\text{N}\cdot\text{m}$ and then from 540 to 600 sec, the torque signal finally levels off at $8.4 \pm 0.3 \mu\text{N}\cdot\text{m}$. Overall, across the 10 min experimental

Figure 6: Representative transient torque response at 11 rad/s during the increasing (filled squares) and decreasing (open squares) vane speed ramps.

time period, the torque signals from the 11 rad/s step during the speed ramp up and ramp down are significantly different from one another and show no sign of converging after long shearing periods (> 5 min). Therefore, the observed hysteresis effect in the wide-gap vane-in-beaker system is not simply a small time-scale dynamic phenomenon and likely arises from different packing histories of the solids bed.

3.3. Torque-Signal Inflection Point

The observed bifurcated torque responses provoke curiosity as to why the inflection points in the torque signals occurred at specific mixing speeds. What do these vane speeds represent and can they be theoretically or empirically predicted? The following section investigates the physical significance of the measured torque-signal inflection point speeds.

In order to further explore the mixing dynamics of the vane, it was first assumed that the vane systems behaved like Couette concentric-cylinders, where the material trapped within the void spaces between the vane blades forms the inner, rotating cylinder (a previously validated assumption [46–48]). Ignoring edge effects, the shear rate ($\dot{\gamma}$) and shear stress (σ) at the vane tip can be approximated as:

$$\dot{\gamma} = \frac{2\Omega}{1 - \kappa^2}, \quad (1)$$

Figure 7: Viscosity ratio (μ_A/μ_N) as a function of Reynolds number (Re) for the viscosity standard oil (open squares) and DI water (filled squares and circles) in the vane-in-beaker (VB) and vane-in-cup (VC) setups. The solid lines are power function fits of the DI water data using the parameters indicated in Table 1.

$$\sigma = \frac{M}{2\pi L R_i^2}, \quad (2)$$

where R_i and R_o are the corresponding vane and vessel radii, $\kappa=R_i/R_o$ is the radius ratio, Ω is the angular speed of the vane, M is the torque response of the vane, and L is the length of the vane's blades. In addition, the Reynolds number, Re, for a Couette concentric-cylinder system can be defined as:

$$\text{Re} = \frac{\Omega R_i R_o (1 - \kappa) \rho}{\mu_N}, \quad (3)$$

where ρ and μ_N are the liquid density and nominal (or reference) viscosity, respectively.

To assess the degree of secondary flows in the vane-in-beaker and vane-in-cup setups, the apparent viscosities of a viscosity oil standard ($\mu_N = 0.1$ Pa·s; Brookfield Engineering Laboratories Inc., Middleboro, Massachusetts, USA) and DI water ($\mu_N = 0.001$ Pa·s [49]) were measured at ~ 20 °C. The standard oil was specifically interrogated in order to acquire data in the low Re regime ($\text{Re} < 100$).

Figure 7 displays the ratio of the apparent viscosity (μ_A , equal to measured shear stress divided by shear rate; see Equations 1 and 2) to the nominal

Table 1: Results from the power function fits (Equation 4).

Vessel	R_i (mm)	R_o (mm)	κ	b	α	R^2
Beaker	7.5	23.5	0.32	0.096	1.56	0.996
Cup	7.5	15.0	0.50	0.063	1.70	0.995
Beaker	14.0	23.5	0.60	0.082	1.65	0.995

viscosity (μ_N) (which will be referred to as the viscosity ratio moving forward) as a function of Re. At low Re (< 50), the viscosity ratio of the standard oil remained as unity (open squares, Figure 7), indicating that turbulent secondary flows were not significant at $\text{Re} < 50$. However, the viscosity ratio of DI water in the vane-in-beaker setups (filled squares and open triangles) and vane-in-cup setup (filled circles) grew exponentially at $\text{Re} > 50$ (Figure 7). The second set of vane-in-beaker data (the third overall experimental setup) was generated with the large, 28 mm vane (measured slurry and DI water torque responses are presented in the Appendix, Figure A.13).

The abrupt and exponential increase in DI water viscosity suggests that there is a critical Re value (Re_C) that signifies the transition between well-defined laminar flow ($\text{Re} < \text{Re}_C$) and the onset of secondary flows ($\text{Re} > \text{Re}_C$). As a result, the DI water viscosity data for the vane-in-cup and vane-in-beaker setups were fit with power functions (solid lines, Figure 7), resulting in expressions that predicted the viscosity ratio as a function of Re:

$$\frac{\mu_A}{\mu_N} = \begin{cases} 1 & \text{if } \text{Re} < \text{Re}_C, \\ b\text{Re}^{\alpha-1} & \text{if } \text{Re} \geq \text{Re}_C, \end{cases} \quad (4)$$

where b and α are constants that can signify the degree of roughness of the inner and outer cylinder walls, as well as describe the energy dissipation rate into the laminar boundary layers and the turbulent bulk in Taylor-Couette flow (valid only under turbulent conditions) [50–52]. The values of α for all three examined experimental setups were between 1.5 to 1.7 (Table 1) and were close to the α value in the smooth cylinder system reported by van den Berg et al. [52], indicating that the laminar boundary layers are still intact and that the vane systems studied here behaved as smooth-Couette concentric cylinders within the examined range of Re.

Using the power function parameters from Table 1, critical Re values (Re_C) for the onset of secondary flows (i.e., when $\mu_A/\mu_N = 1$) were obtained for each experimental setup (Table 2). The empirically predicted Re_C values

336 ranged from 48 to 64, which are relatively small. Work by Andereck et al. [53]
 337 graphically presented the transitions in flow regimes for two generic rotating
 338 concentric cylinders via visual observations. When the outer cylinder was
 339 stationary, the flow transition boundaries were roughly defined as: azimuthal
 340 Couette flow: $Re < 100$, Taylor-vortex flow: $100 < Re < 150$, Wavy vortex
 341 flow: $150 < Re < 1000$, and turbulent flow: $Re > 1000$. According to the
 342 flow regime definitions by Andereck et al., the empirically determined Re_C
 343 values from Table 2 would reside in the laminar (or Couette) flow regime.
 344 However, more recent work by Sprague et al. [54] presented critical Reynolds
 345 numbers for the onset of Taylor vortices as a function of radius ratio (κ)
 346 for Couette cells with infinitely long annuluses. Theoretically computed Re_C
 347 values (extracted from Figure 2 in Sprague et al. [54]) ranged from 68 to 73
 348 and are in agreement with the empirically defined Re_C values (Table 2).

349 Using the critical Re values (both empirical and theoretical) and a nomi-
 350 nal slurry viscosity of 0.017 Pa·s (5 wt% α -cellulose; Appendix, Figure A.11),
 351 critical vane speeds (Ω_C) were calculated for each experimental system using
 352 Equation 3. The empirically and theoretically derived Ω_C values represent
 353 speeds in which secondary flows exist that can potentially keep the parti-
 354 cles in suspension. These empirical and theoretical values of Ω_C were then
 355 compared to experimental critical vane rotation rates during the increasing
 356 and decreasing speed ramps, $\Omega_{C,IS}$ and $\Omega_{C,DS}$, respectively (Table 2). For
 357 all three experimental setups, the predicted critical vane rotation rates were
 358 similar to the measured range of Ω_C values. Due to the abnormal drop in the
 359 torque signal during the increasing speed ramp in the 15 mm vane-in-beaker
 360 system ($\kappa = 0.32$) no measured Ω_C was reported. The empirical and theo-
 361 retical predictions not only agreed with experimental values, but they also
 362 coincided with one another, which suggests that the observed bifurcations
 363 in the torque signals were due to the onset of secondary flows (specifically
 364 Taylor vortices).

365 Overall, the empirical predictions (corroborated with theoretical com-
 366 putations [54]) indicate that secondary flows are necessary in order to fully
 367 resuspend an initially settled bed of fibrous, non-colloidal particles. By defin-
 368 ing the onset of secondary flows using empirical fits of data from ideal fluids
 369 (i.e., DI water), allowed for unique critical Re values to be predicted for each
 370 experimental setup. The Re_C values, in conjunction with the nominal slurry
 371 viscosity, were then used to effectively predict a critical mixing speed for each
 372 experimental system.

Table 2: Comparison of empirically and theoretically predicted critical Reynolds numbers (Re_C) and calculated mixing speeds (Ω_C) with experimentally measured Ω_C values (the subscripts IS and DS indicate the increasing and decreasing speed ramps, respectively).

Vessel	κ (-)	Empirical [†]		Theoretical ^{††}		Experimental	
		Re_C (-)	Ω_C (rad/s)	Re_C (-)	Ω_C (rad/s)	$\Omega_{C,IS}$ (rad/s)	$\Omega_{C,DS}$ (rad/s)
Beaker	0.32	64	8.7	73	10.4	- *	13
Cup	0.50	52	15.1	68	20.7	15	15
Beaker	0.60	48	5.9	72	9.2	9	9

[†] Empirical Re_C values were obtained from fits of the DI water data in Figure 7.

^{††} Theoretical Re_C values were extracted from Figure 2 in Sprague et al. [54].

* No critical mixing speed was reported during the increasing speed ramp due to the irregular torque response.

3.4. Torque Response Prediction

We now consider the prediction of the entire torque signal during slurry settling and resuspension. Assuming Newton’s law of viscosity holds true and the flow is laminar and purely azimuthal, then Equations 1 and 2 can be combined and rearranged to obtain an expression for the torque response of the vane (M):

$$M = \frac{4\pi\mu L\Omega R_i^2}{1 - \kappa^2}, \quad (5)$$

where μ is the material’s viscosity. However, the system of interest for this study is a two-phase mixture that only behaves as a single-phase continuum at high shear speeds. When the system is in the “settled regime” and not well mixed, one must account for the liquid-phase resistance (μ_L) and the settled-solids-bed resistance (μ_S), separately. However, μ_L and μ_S are not constant values and in addition, they are highly dependent on the fraction of solids in suspension versus the amount of solids in the settled bed. Therefore, for this system, the fraction of settled-to-suspended solids (ε) can be defined as the ratio of the solids bed height, h (minus the 4 mm spacing between the bottom of the vessel and the vane), to the vane blade length, L . Therefore, the total system effective viscosity (μ_T) can be expressed as:

$$\mu_T = (1 - \varepsilon)\mu_L + \varepsilon\mu_S, \quad (6)$$

where the μ in Equation 5 can be replaced by μ_T .

Using images (a) and (b) from Figure 5 ($h_a=22$ mm and $h_b=16$ mm) and assuming that solids are resuspended at the same rate in both systems and across all vane speeds, an expression for the reduction in ε as a function of vane rotation rate (Ω) can be approximated:

$$\varepsilon = -0.038\Omega + 0.69 \text{ (vane-in-cup)}, \quad (7)$$

$$\varepsilon = -0.038\Omega + 0.51 \text{ (vane-in-beaker)}, \quad (8)$$

where the intercept for each setup is equal to ε_0 : the ratio of the maximum, undisturbed solids bed height (h_0) to the vane blade length (L). The cup was filled with sample to a slightly higher fraction than the beaker, so at equivalent solids loadings and spacings between the vane and bottom of the vessel, ε_0 was larger for the vane-in-cup setup. Additional images from Figure 5 were not used in the development of Equations 7 and 8 because at rotation rates above 5 rad/s, particles began to settle out of suspension near the beaker wall and the observed solids bed height actually increased.

In addition, μ_S and μ_L are functions of ε . Using the initial settled solids bed height h_0 and vessel dimensions, the weight fraction of solids in the settled bed was estimated to be ~ 0.103 for both systems. A slurry with 10.3 wt% solids has an approximate viscosity of 3.0 Pa·s (Appendix, Figure A.11), which was the value used as the initial estimate of μ_S . Also, not only are particles being resuspended as the vane rotation rate increases (reducing ε), but the solids bed is also becoming less rigid. As the vane is slicing through the compacted solids bed, particles will reposition and lose contact in order to slide past one another and flow. The loosening of the particle bed will make the settled solids more “fluidized” and reduce its viscosity (μ_S). Therefore, μ_S will decrease with increasing vane rotation rate. The reduction of μ_S with increasing vane speed (Ω) was defined for both systems as:

$$\mu_S = C_S \Omega^{-1.35}, \quad (9)$$

where $C_S = 3.0$ Pa·s. Equation 9 was derived using a power-law fit of the two known bounding states of the slurry: 1) Fully settled: $\Omega = 1$ rad/s, where $\mu_S = 3.0$ Pa·s and 2) Fully suspended: $\Omega = 45$ rad/s (the maximum experimentally examined vane rotation rate), where $\mu_S = \mu_L = 0.017$ Pa·s (previous observations or knowledge of when the solids were “actually” fully suspended were ignored).

When all of the particles are settled out of suspension ($\varepsilon = \varepsilon_0$), μ_L will equal the viscosity of water (0.001 Pa·s at 20 °C). Conversely, when all of

the particles are fully suspended ($\varepsilon = 0$), μ_S is negligible, and μ_L will be equivalent to the viscosity of a fully suspended 5 wt% α -cellulose slurry (μ_{SL}), where $\mu_{SL} = 0.017$ Pa·s (until turbulence plays a role, see below). Using the vessel dimensions and Equations 7 and 8, the weight fraction of suspended solids (x_s) can be estimated and correlated with ε . Then the model fit which predicts slurry viscosity for a given value of x_s (Appendix, Figure A.11) can be used to determine μ_L as a function of ε for each experimental setup:

$$\mu_L = \mu_{SL} e^{-4.0\varepsilon} \text{ (vane-in-cup), } \text{Re} < \text{Re}_C, \quad (10)$$

$$\mu_L = \mu_{SL} e^{-5.5\varepsilon} \text{ (vane-in-beaker), } \text{Re} < \text{Re}_C. \quad (11)$$

Since the resuspension of solids was assumed to be equivalent for both scenarios and ε_0 is larger for the vane-in-cup system, it takes longer for ε to reduce to zero using the cup (hence the slightly smaller exponent in Equation 10). For mixing above the critical mixing rate in a specified system,

$$\mu_L = \mu_{SL} b(\text{Re})^{\alpha-1}, \text{Re} \geq \text{Re}_C, \quad (12)$$

where α and b are system-dependent empirical coefficients, taken from Table 1.

The empirical relationships for ϵ , μ_S , μ_L , and μ_T in the vane-in-cup system are shown graphically in Figure 8. Initially from 1 to 5 rad/s, μ_T is strongly correlated with μ_S , as μ_L is too small to make a large impact on the value of μ_T . However, at vane rotation rates above 10 rad/s, μ_L becomes large enough to influence μ_T and at vane speeds > 15 rad/s, μ_L and μ_T are equivalent.

Using the above expressions to calculate μ_T , the torque response of the vane rotor during resuspension of a fully settled 5 wt% α -cellulose slurry can now be predicted. For the vane-in-cup setup, the model mimics the experimental data well (filled circles, Figure 9); predicting an inflection point in the torque signal at ~ 16 rad/s, which is comparable to the measured inflection point speed for both the increasing and decreasing speed ramps. In the settled regime, the model and measured torque values are in close agreement. The systematic over-prediction of the torque response by the model in the suspended regime (which continues to diverge with increasing speed) indicates that either the nominal viscosity of the 5 wt% α -cellulose slurry was actually less than what was independently measured (Appendix, Figure A.11), the viscosity ratio correlation over-estimates the degree of turbulent secondary flows, or perhaps the centripetal force from the vane is driving

Figure 8: (Left axis) Predicted total system effective viscosity (μ_T , red circles) and solids bed (μ_S , open circles) and liquid (μ_L , gray circles) contributions to μ_T as a function of rotation rate (Ω) for the vane-in-cup setup. (Right axis) The ratio of the settled bed height to the vane blade length (ε , blue squares) as a function of Ω .

particles away from the vane in the radial direction (i.e., shear-induced migration). In addition, the governing expressions used to develop the torque model (Equations 1 - 3 and 5), assume 2D Taylor-Couette flow and that the vane behaves as a solid cylinder. However, these assumptions do not perfectly describe the vane's shear profile, for example: distorted stream-lines, edge effects, slip at the outer vessel wall, and vortices forming behind the blades (to name a few) can potentially impact the torque response of the vane (especially when inertial effects become important) [47, 48, 55].

The predicted torque is shown along with the experimental data for vane-in-beaker system in Figure 10. Although the predicted torques are systematically larger than the experimental data, the predictions are closer for the decreasing-ramp data. The discrepancy between the model and experiment during the increasing speed ramp is most likely a direct result of the model not accounting for the shear-induced transport of particles from areas of high shear (the vane) to areas of low shear (the vessel wall); which was evident during experimentation in the vane-in-beaker system (Figure 5).

In general, the presented torque model provides qualitative agreement with the results from both experimental systems (vane-in-cup and vane-in-beaker), and in some cases semi-quantitative agreement is achieved. The

Figure 9: Predicted torque response of a 5 wt% α -cellulose slurry (with and without settling) in the vane-in-cup setup ($\kappa = 0.50$) as a function of angular speed (or Re). The measured torque values during the increasing (filled triangles) and decreasing (open triangles) speed ramps are from Figure 3.

Figure 10: Predicted torque response of a 5 wt% α -cellulose slurry (with and without settling) in the vane-in-beaker setup ($\kappa = 0.32$) as a function of angular speed (or Re). The measured torque values during the increasing (filled squares) and decreasing (open squares) speed ramps are from Figure 4.

475 vane-in-beaker is a non-ideal rheological system that better mimics a true
 476 mixing vessel and as a result, it presents additional challenges when attempt-
 477 ing to describe its torsional response during resuspension. Therefore, a more
 478 detailed model beyond our simplistic representation is needed in order to
 479 fully represent the resuspension of a completely settled particle bed in the
 480 wide-gap vane-in-beaker system. However, the critical length relationship
 481 when describing the resuspension of settled solids appears to be related to
 482 the radius ratio (κ) of the mixing system (as indicated in Section 3.3), as
 483 well as the ratio of the solids bed height to the penetration depth of vane
 484 rotor (i.e., ε).

485 4. Conclusions

486 The 5 wt% α -cellulose slurry displayed a diverged torque response as a
 487 function of increasing and decreasing vane mixing speed, clearly defining re-
 488 gions where settling and suspension were dominant. At low rotational rates,
 489 where Re is less than Re_C , the inertial forces from the rotating vane were
 490 too weak to keep the particles fully suspended, resulting in an increased
 491 torque signal (i.e., a “settled” regime develops). However, at relatively large
 492 vane speeds, where Re is greater than Re_C , mixing forces become strong
 493 enough to keep the α -cellulose particles suspended and the slurry displays
 494 Newtonian-like flow behavior. In addition, when using the vane-in-beaker
 495 setup, a hysteresis was observed. Particles were deposited near the beaker
 496 wall during the increasing speed ramp and the region around the vane be-
 497 came devoid of particles, registering a torque signal from the slurry that was
 498 equivalent to that of DI water. The observed hysteresis is believed to be
 499 a result of the large gap between the vane and beaker wall, as well as the
 500 different compaction histories of the settled solids.

501 In addition, the torque response of model fluids (namely DI water) were
 502 used to empirically define critical Reynolds numbers at which sufficient mix-
 503 ing turbulence was established in each experimental setup. Based on this
 504 Re_C value, critical vane mixing speeds were predicted. The predicted critical
 505 mixing speeds were in strong agreement with measured values, indicating
 506 that secondary flows (specifically Taylor vortices) are required in order to
 507 keep the cellulose particles fully suspended. This approach has the potential
 508 to be applied to real-world mixing systems, where proper mixing speeds for
 509 difficult-to-characterize, multi-phase materials (like biomass slurries) can be
 510 inferred empirically from the torque response of ideal fluids. Also, a simplistic

511 scaling model was developed using experimental parameters that predicted
 512 the torque response of a slurry during settling and resuspension. The model
 513 generally agreed with the experimental results from both setups, but was
 514 unable to capture the strong radial shear-induced transport effects during
 515 resuspension in the vane-in-beaker system. However, all of the presented
 516 diagnostic approaches (direct experimental interrogation, empirical predic-
 517 tion, or torque modeling), alone or in combination, should help engineers
 518 better understand the mixing conditions and torque requirements necessary
 519 for achieving a well-mixed dispersion of negatively buoyant biomass particles.

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Appendix A. Additional Figures

The viscosities of α -cellulose slurries with varying solids weight fractions were measured using a Bohlin Gemini HR Nano stress controlled rheometer (Malvern Instruments, Malvern, UK). In order to combat shear-induced particle migration and help limit the effects of settling, slurry viscosity was probed using roughened, 40 mm parallel-plates. The rough plates were machined with surface serrations of ~ 1 mm, which grip the cellulose particles and help eliminate slip at the sample-tool interface. Using a gap height of $1000\ \mu\text{m}$, shear rates ranging from 1 to $1000\ \text{s}^{-1}$ were examined (shear-induced particle migration typically occurs at shear rates well above $1000\ \text{s}^{-1}$ in the parallel-plate geometry [56]). Some measurements were repeated in duplicate and the general scatter of the data provides a qualitative assessment of measurement reproducibility (all data are reported in Figure A.11).

732 As expected, the slurry’s viscosity decreased with increasing shear rate
 733 (i.e., shear thinned) until reaching a Newtonian plateau at elevated strain
 734 rates ($>10 \text{ s}^{-1}$), indicating the slurry was fully suspended. Figure A.11
 735 shows a plot of the slurry’s Newtonian viscosity as a function of α -cellulose
 736 solids concentration. As anticipated, the constant, high shear viscosity of
 737 the α -cellulose slurry increased with increasing solids fraction (Figure A.11).
 738 The slurry viscosity data were fit using a modified Krieger-Dougherty model
 739 [57], which predicts slurry viscosity as a function of solids weight fraction
 740 (the model and fit parameters are displayed in Figure A.11). The model was
 741 used to determine the “nominal” α -cellulose slurry viscosity.

742 The empirical relationships for ϵ , μ_S , μ_L , and μ_T for the vane-in-beaker
 743 setup are shown in Figure A.12. Initially from 1 to 5 rad/s, μ_T is strongly
 744 correlated with μ_S , as μ_L is not large enough to significantly impact the value
 745 of μ_T . However, at vane rotation rates above 10 rad/s, the contribution of
 746 μ_L becomes strong enough to influence μ_T , and at vane speeds $> 13 \text{ rad/s}$,
 747 μ_L and μ_T are equal.

748 The 5 wt% α -cellulose slurry displays a bifurcated torque response with
 749 increasing and decreasing angular speed in the 28 mm vane-in-cup setup
 750 (Figure A.13). The inflection point in the torque signal (i.e., the rotation
 751 rate that signifies the transition between the settled and suspended regimes)
 752 was observed at 9 rad/s. Figure A.13 also shows the steady torque response
 753 of DI water and the model prediction for a fully suspended slurry devoid of
 754 settling.

Figure A.11: Measured viscosity of α -cellulose slurries with varying solids weight fractions (filled circles). The line represents a model (expression displayed in figure) that predicts slurry viscosity (μ) with a known solids weight fraction (x_s). The model was used to determine the nominal slurry viscosity (μ_N) used within the manuscript.

Figure A.12: (Left axis) Predicted total system effective viscosity (μ_T , blue circles) and solids bed (μ_S , open circles) and liquid (μ_L , gray circles) contributions to μ_T as a function of angular speed (Ω) for the vane-in-beaker setup. (Right axis) The ratio of the settled bed height to the vane blade length (ε , red squares) as a function of Ω .

Figure A.13: Steady torque as a function of increasing (filled squares) and decreasing (open squares) vane rotation rate (or Re) for an initially fully settled (16 hrs) 5 wt% α -cellulose slurry in the 28 mm vane-in-beaker setup. The torque response of the slurry is compared to the torque signal of the DI water control (filled circles) and the model prediction for a fully suspended slurry in the absence of settling (open circles).