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Particle agglomeration studies in a slurry bubble column due to liquid bridging: Effects of particle size and sparger design

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HIGHLIGHTS

- Overall and axial phase holdups in a slurry bubble column were measured.
- An increase in secondary liquid phase viscosity resulted in greater sedimentation.
- Larger particles sedimented at lower secondary liquid phase loadings.
- Three sparger designs were tested for their impact on sedimentation mitigation.
- Revolution Powder Analyzer measurements were compared to the slurry bubble column.

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ABSTRACT

Particle agglomeration can occur in heavy oil upgrading hydroprocessors due to the formation of carbonaceous mesophase, a secondary liquid phase which results from an increased rate of thermal cracking relative to the hydrogenation rate. A cold-flow slurry bubble column operated at atmospheric pressure with an internal diameter of 0.152 m was used to examine particle agglomeration behavior and the overall fluid dynamics in a gas-liquid-liquid-solid system. The experimental system consisted of biodiesel (organic continuous phase), aqueous glycerol solutions (immiscible secondary phase), glass beads and nitrogen. The impacts of the secondary liquid loading, secondary liquid viscosity, and particle diameter on the fluid dynamics were established. Particle agglomeration was studied by measuring the axial solid holdup profiles while varying the superficial gas velocity and secondary liquid loading. Enhanced particle agglomeration was observed when increasing the secondary liquid loading, based on the increased solid holdups at the bottom of the column. Three sparger designs (six-legged spider sparger, perforated plate and conical perforate plate) were compared. Following the glycerol addition, the spider sparger was less effective based on the particle sedimentation at lower liquid/solid ratios when compared to the other sparger designs. A Revolution Powder Analyzer (RPA) was also used for complementary measurements to examine whether operational difficulties due to particle agglomeration can be anticipated using a simplified system.

1. Introduction

Particle agglomeration in gas-liquid-liquid-solid (G-L-L-S) fluidized beds can be caused by Van der Waals forces, electrostatic/Coulombic forces and/or interparticle liquid bridging. Van der Waals forces are attractive forces that occur due to interaction between permanent dipoles (Keesom forces), permanent/induced dipoles and dispersion forces of non-polar molecules (London dispersion forces). Van der Waals forces generally affect particles in the micron range (smaller than 10 μm) (Simons, 1996). Electrostatic attractive forces occur due to interactions between particles with opposite charges, which may be caused by triboelectricifaction, ion collection, thermionic emission or frictional charging (Park and Fan, 2007). These forces typically affect particles with low electrical dissipation rates, such as polymers (Park et al., 2002; Sowinski et al., 2010). The use of a liquid in G-L-S or G-L-L-S fluidized beds generally increases the system's electrical dissipation, reducing electrostatic forces between particles. Lastly, liquid bridges are formed when two particles, surrounded by a layer of a wetting liquid, collide and form a bridge, binding the

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Nomenclature

a	dimensionless number from the relation from Cheng (2008)
b	dimensionless number from the relation from Cheng (2008)
C_m	glycerol mass fraction in an aqueous glycerol mixture (w/w)
d_C	column inner diameter (m)
d_O	orifice diameter (m)
d_p	average particle diameter (m)
e	coefficient of restitution (–)
g	gravitational acceleration (m/s^2)
h	binder layer thickness covering colliding granules (μm)
h_0	particle half-gap distance (μm)
H_E	expanded bed height (m)
H_i	initial bed height (m)
K_d	gas sparger coefficient from Behkish et al. (2006)
L/S	volumetric secondary liquid-to-solid ratio (–)
m	granule mass (g)
N_O	number of orifices in the gas sparger
P	pressure (Pa)
ΔP	dynamic pressure drop (Pa)
P_S	vapor pressure of the liquid (MPa)
P_T	total pressure (MPa)
PfP	perforated plate sparger
St_V	viscous Stokes number
St_V^*	critical viscous Stokes number
T	temperature (°C)
u_0	initial relative granule collisional velocity (cm/s)
U_G	gas superficial velocities (m/s)
x	mass fraction (w/w)
X_W	weight fraction of the primary liquid in the mixture ($1 \geq X_W \geq 0.5$) (w/w)
We_G	Weber number
z	vertical distance (m)

Greek symbols

α	weighting factor varying from 0 to 1 from the relation from Cheng (2008)
ε_G, ε_L, ε_S	gas, liquid and solid holdups in the bed region
Γ	gas distributor parameter used in Eq. (6)
μ_g, μ_w	glycerol and water viscosity (Pa·s)
$\mu_{L,C}$, $\mu_{L,I}$	continuous and secondary liquid viscosity (Pa·s)
ξ	sparger to column cross sectional area ratio in Eq. (6)
ρ_{GB}	glass bead density (kg/m^3)
$\rho_{L,C}$, $\rho_{L,I}$	continuous and secondary liquid densities (kg/m^3)
ρ_G, ρ_S	gas and solid densities (kg/m^3)
$\sigma_{L,C}$	continuous liquid surface tension (N/m)
ω	fraction of the total packed bed voidage where the total voidage is 1.00
ψ	gas-free overall solid holdup

Subscripts

C	continuous
I	secondary
G	gas
GB	glass beads
L	liquid
S	solid

particles together. Particle agglomeration in a nitrogen-biodiesel-glass bead system has been previously observed following the addition of a secondary immiscible liquid, such as glycerol (Pjontek et al., 2014), which wets the particles. The attractive force for the previous system is thus interparticle liquid bridging, providing a G-L-L-S configuration with particle agglomeration due to liquid bridging in a slurry bubble column.

Hydroprocessing a heavy feedstock can generate mesophase, a secondary liquid phase, due to an accelerated rate of thermal cracking relative to the hydrogenation rate. Hydroprocessing is a temperature-sensitive process, whereby small temperature increases can result in the formation of the previous coke-precursor material. Mesophase was first identified and characterized by its optical anisotropy when observed under polarized light (Bisoyi and Kumar, 2010; Brooks and Taylor, 1965). Coke is generally defined as toluene insoluble materials and is believed to originate from the asphaltene fraction in the hydroprocessor feedstock (Srinivasan and McKnight, 1994). Mesophase is thus an intermediate phase between solid coke and vacuum residue, and is characterized as polar, denser and more viscous when compared to the continuous liquid in a hydroprocessor (Srinivasan and McKnight, 1994). Previous studies have discussed potential formation mechanisms (Bagheri et al., 2012; Gray and McCaffrey, 2002; Wiehe, 1994). A recent study by Bagheri et al. (2012) investigated the in-situ formation of mesophase in a stirred hot-stage reactor (440 °C and 4.8 MPa). The study noted the presence of large (surface area greater than 2000 μm^2) and small (surface area less than 2000 μm^2) mesophase domains and suggested that the large domains were formed due to the coalescence of smaller domains. Another study by Sharshar et al. (2015) studied the effects of temperature, catalyst concentration, partial pressure of hydrogen and agitation speed on the mesophase onset time. The authors found that the mesophase onset time decreased at higher temperatures and lower catalyst concentrations. The hydrogen partial pressure and agitation speed were not found to have a significant impact on mesophase onset time for the studied operating conditions.

The formation of this secondary immiscible liquid phase may result in particle agglomeration due to liquid bridging and could impact the fluidized bed behavior. Previous studies have examined the effects of particle size, shape and material in a cold-flow ebullated bed (Pjontek et al., 2011, 2014) and the secondary liquid phase loading in a slurry bubble column (Siquier et al., 1991). Pjontek et al. (2014) studied a nitrogen-biodiesel-glycerol system with glass beds and aluminum cylinders. The study noted that the smaller diameter particles (d_p = 1.5 mm) demonstrated more clustering when compared to the larger particles (d_p = 4 mm). The authors also carried out preliminary measurements in a slurry bubble column and found that the G-L-L-S system with glass beads between 100 and 150 μm was inoperable at a much lower glycerol loading (0.7 wt.% overall liquid loading) when compared to the ebullated bed, which was still operable at a glycerol loading of 5 wt.%. Siquier et al. (1991) measured the density gradient using a sampling probe as a secondary liquid was added to the system in a cold-flow atmospheric slurry bubble column using atmospheric air, kerosene, water and glass beads (d_p = 110 μm). The authors identified three different situations depending on the operating conditions:

- **Case 1:** Two regions of solid suspension are observed. At the bottom of the column, all four phases are present and particles have begun to cluster due to the secondary immiscible liquid. Above this region, there are small amounts of particles suspended in the continuous liquid and no secondary phase is present.