An investigation into the influence of particle properties and operating conditions on gas-solid flow phenomena

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**NANYANG
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SINGAPORE

**AN INVESTIGATION INTO THE INFLUENCE OF
PARTICLE PROPERTIES AND OPERATING
CONDITIONS ON GAS-SOLID FLOW PHENOMENA**

**ADITYA ANANTHARAMAN
SCHOOL OF CHEMICAL AND BIOMEDICAL ENGINEERING
2017**

An Investigation into the Influence of Particle Properties and Operating Conditions on Gas-Solid Flow Phenomena

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Abstract

Horizontal pneumatic conveying and fluidization are two important gas-solid flow operations employed in wide-ranging industries from agro-processing to pharmaceutical and oil and gas. Despite being employed in industry for decades, they are still largely run on rules-of-thumb and experience rather than scientific principles. This research effort endeavors to contribute to their physical understanding by analyzing the effects of particle properties and varying operating conditions on these phenomena. This thesis is divided into three parts focusing on horizontal pneumatic conveying, fluidization and reactor modeling respectively.

Part I deals with the minimum pickup velocity (U_{pu}), defined as the minimum gas velocity required to initiate motion of a particle initially at rest, a very important parameter in horizontal pneumatic conveying. In this section, firstly, the effect of continuous particle size distribution (PSD) and particle shape on U_{pu} is studied, where the involvement of inter-particle momentum transfer and particle rotation and lift is revealed. Secondly, the effect of particle diameter, density and shape on U_{pu} is studied by investigating binary mixtures. Thirdly, the U_{pu} of nanoparticles is reported for the first time and behavioral differences between polar and apolar nanoparticles are noted. Finally, the knowledge gap in U_{pu} at the nano- and micro- scales is bridged by investigating particles from diameters 5 nm to 110,000 nm and the Three-zone Model developed for the micro-scale is modified to incorporate nanoparticles.

Part II discusses the minimum fluidization velocity (U_{mf}), and differential pressure signals and radial mass flux distribution in circulating fluidized beds (CFBs). Specific studies include the following: (i) a comparative analysis of the predictions of over a hundred empirical correlations to determine U_{mf} with respect to Geldart Groups A, B and D, and bed voidage and particle sphericity; and (ii) investigating the effects of particle properties, namely, particle diameter and density, and varying operating conditions, namely, superficial gas velocity and

overall mass flux on differential pressure signals (via Discrete Fourier Transform and Wavelet Decomposition) and the radial mass flux behavior of particles in a pilot-scale CFB riser.

Part III attempts numerical modeling of a carbon-dioxide (CO₂) methanation bubbling fluidized bed reactor. The effects of operating conditions like the temperature, pressure, and inlet feed ratio, and hydrodynamic parameters like the inlet feed rate, superficial gas velocity, particle diameter, particle density, particle sphericity and the bed voidage at minimum fluidization on methane production by the Sabatier reaction are studied through a heterogeneous bubbling fluidized bed reactor model.

The physical understanding on horizontal pneumatic conveying and fluidization presented in this thesis is expected to contribute to the database and aid future researchers as well as designers of systems employing these operations.

DEDICATION

To

Appa, Mamma, Anuja and Dada

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Nomenclature

A	Projected area of a particle (m^2) (Chapters 3 and 4)
	Empirical constant in a correlation to predict U_{mf} (-) (Chapter 7)
	Cross-sectional area of the reactor (m^2) (Chapter 11)
a	Specific mass-transfer area (m^{-1})
A_H	Hamaker coefficient (J)
$A_j(t)$	Approximation signal at the j th scale in wavelet decomposition
A_p	Projected area of the modeled non-spherical particle (m^2)
Ar	Archimedes number (-)
Ar^*	Archimedes number modified for particle sphericity (-)
b	Empirical constant in a correlation to predict U_{mf} (-)
C_1, C_2	Empirical constants in a correlation to predict U_{mf} (-)
C_D	Drag coefficient (-)
D	Pipe diameter (m)
	Riser diameter (m)
d^*	Diameter of primary nanoparticle agglomerate (μm (Chapter 5), nm (Chapter 6))
d^{**}	Diameter of complex nanoparticle agglomerate (μm (Chapter 5), nm (Chapter 6))
D_0	Fractal dimension of primary nanoparticle agglomerate (-)
D_1	Fractal dimension of complex nanoparticle agglomerate (-)
d_{ave}	Average particle diameter (m (unless otherwise specified))
d_b	Bubble diameter (m)
d_{b0}	Initial bubble diameter, 0.00853 m
D_{cmin}	Diameter of the minimum circumscribed circle around the projected 2-D image of the particle (m)
$D_{i,j}$	Binary diffusion coefficient of species i and j ($\text{m}^2 \text{s}^{-1}$)
$D_{i,mix}$	Molecular diffusion coefficient of species i ($\text{m}^2 \text{s}^{-1}$)
$D_j(t)$	Detail signal at the j th scale in wavelet decomposition
d_p	Particle diameter (m (unless otherwise specified))
$d_{p,mix}$	Characteristic particle diameter of a binary mixture (m)
d_{p1}, d_{p2}	Particle diameters of the two constituent species of a binary mixture (m)

d_{p50}	Median particle diameter (μm)
D_{ref} or D_{50}	Reference pipe diameter (0.05 m)
d_{sm}	Sauter-mean diameter (μm)
E_{Dj}	Energy of a detail signal at decomposition scale j
$E_{Dj,empty}$	E_{Dj} when $U_s = 0$ and $G_s = 0$
f	Coefficient of friction (-)
f	Signal frequency (Hz)
$f1(\theta), f2(\theta)$	Functions of θ used in the expression to calculate minimum pickup velocity (-)
F_b	Buoyancy force (N)
F_D	Drag force acting on a particle (N)
F_f	Frictional force (N)
F_g	Gravitational force (N)
f_m	Mass-based frequency distribution (μm^{-1})
F_{OH}	Force of attraction due to hydrogen bonds (N)
F_{vdw}	Van der Waal force of attraction (N)
g	Acceleration due to gravity (9.81 m s^{-2})
$G_{r,downward}$	Downward local solids mass flux ($\text{kg m}^{-2}\text{s}^{-1}$)
$G_{r,net,norm}$	Normalized radial solids mass flux (-)
$G_{r,upward}$	Upward local solids mass flux ($\text{kg m}^{-2}\text{s}^{-1}$)
G_s	Overall solids mass flux ($\text{kg m}^{-2}\text{s}^{-1}$)
GSD	Geometric standard deviation (-)
H	CFB Riser height (m)
h	Height (along the CFB riser) evaluated (m)
j	Imaginary unit (Chapter 10)
K	Empirical constant in a correlation to predict U_{mf} (-)
k	Reaction constant ($\text{mol kg}^{-1}\text{s}^{-1}\text{kPa}^{-5/6}$)
k_0, k_1	Coefficients in a two-level hierarchical fractal consisting of primary and complex agglomerates (-)
K_1, K_2	Empirical constants in a correlation to predict U_{mf} (-)
$K_{G,i}$	Inter-phase mass transfer coefficient of species i due to concentration gradient (m s^{-1})
K_i	Adsorption constant of species i (units differ, see Eqs. (11.23-11.25))

m	Solids loading ratio (-)
M_i, M_j	Molecular weight of species i and j , respectively (kg mol^{-1})
N	Fitting parameter to include the connectivity of the primary nanoparticle agglomerates (-) (Chapter 5)
	Number of discrete points in the timeseries signal $x(t)$ (-) (Chapter 9)
n	Coefficient to account for flow over a layer of particles (-) (Chapter 4)
	Number of correlations in the category for which the GSD is being determined (-) (Chapter 7)
N_{vc}	Convective inter-phase mass transfer ($\text{mol m}^{-1} \text{s}^{-1}$)
P	Pressure (Pa)
p	Empirical constant in a correlation to predict U_{mf} (-)
P_i	Partial pressure of species i (Pa)
q	Empirical constant in a correlation to predict U_{mf} (-)
R	Radius of the sphere which is cut along a segment to model a non-spherical particle (m) (Chapter 4)
	CFB Riser radius (m) (Chapters 8, 9 and 10)
	Ideal gas constant ($\text{m}^3 \text{Pa K}^{-1} \text{mol}^{-1}$) (Chapter 11)
r	Radial position evaluated in a CFB riser (m)
	Reaction rate ($\text{mol kg}_{\text{cat}}^{-1} \text{s}^{-1}$) (Chapter 11)
R^2	Coefficient of regression (-)
Re_{DPF}	Threshold Reynolds number for dense phase flow (-)
Re_{DSU}	Threshold Reynolds number for dense suspension upflow (-)
Re_{mf}	Reynolds number at minimum fluidization (-)
Re_p	Particle Reynolds number (-)
Re_p^*	Modified particle Reynolds number (-)
R_i	Rate of consumption or formation of component i ($\text{mol kg}_{\text{cat}}^{-1} \text{s}^{-1}$)
St	Stokes number (-)
t	Time (s)
T	Temperature (K)
U, U_g	Superficial gas velocity (m s^{-1})
U_b	Bubble velocity (m s^{-1})
U_{im}	Incipient motion velocity (m s^{-1})
U_{mf}	Minimum fluidization velocity (m s^{-1})

$U_{mf,i}$	Minimum fluidization velocity predicted at a d_p by a correlation of the category for which GSD is being determined (m s^{-1})
U_{pu}	Minimum pickup velocity (m s^{-1})
$U_{pu,50}$	Minimum pickup velocity normalized for a channel inner diameter of 50 mm (m s^{-1})
$U_{pu,calculated}$	Calculated minimum pickup velocity (m s^{-1})
$U_{pu,less\ dense}$	Minimum pickup velocity of that constituent species of a binary mixture having the lower particle density (m s^{-1})
$U_{pu,lower}$	Minimum pickup velocity of that constituent species of a binary mixture having the lower minimum pickup velocity (m s^{-1})
$U_{pu,measured}$	Experimentally measured minimum pickup velocity (m s^{-1})
$U_{pu,small}$	Minimum pickup velocity of that constituent species of a binary mixture having the lower particle diameter (m s^{-1})
U_{salt}	Minimum saltation velocity (m s^{-1})
V	Volume of modeled non-spherical particle (m^3) (Chapter 4) Total volume of the reactor (m^3) (Chapter 11)
v_b	Visible bubble phase velocity based on tube diameter (m s^{-1})
V_c	Volume of the portion that is cut-off from the sphere to model a non-spherical particle (m^3)
V_{ca}	Type A choking velocity (m s^{-1})
v_i	Stoichiometric factor of species i (-)
V_{mp}	Minimum pressure velocity (m s^{-1})
v_s	Cross-sectional average particle velocity (m s^{-1})
V_{sph}	Volume of entire sphere used to model a non-spherical particle (m^3)
v_t	Particle terminal velocity (m s^{-1})
X	$\left(\frac{d_p^2 (\rho_p - \rho) g}{\mu} \left(\frac{\rho_p}{\rho} \right)^{1.23} \right) (\text{m s}^{-1})^{(1/\alpha)}$
$X(f)$	Discrete Fourier Transform (DFT)
$x(t)$	Time series signal
x_1, x_2	Mass fractions of the constituent species of a binary mixture with particle diameters of d_{p1} and d_{p2} or particle densities of ρ_{p1} and ρ_{p2} or particle sphericities of ϕ_1 and ϕ_2 respectively (-)
$x_{b,i}$	Mole fraction of component i in the bubble phase (-)

x_i, x_j	Mole fraction of species i and j , respectively (-)
$y_{b,i}$	Bubble phase concentration of species i (mol m ⁻³)
$y_{b,i,feed}$	Bubble phase concentration of species i at reactor inlet ($z = 0$) (mol m ⁻³)
$y_{e,i}$	Emulsion phase concentration of species i (mol m ⁻³)
$y_{e,i,feed}$	Emulsion phase concentration of species i at reactor inlet ($z = 0$) (mol m ⁻³)
z	Axial position along the fluidized bed column (m)
z_{eq}	Equivalent distance between two nanoparticle agglomerates (m)
α	Parameter proportional to strength of the hydrogen bond, concentration of hydroxyl groups on the nanoparticle agglomerate surface, fraction of agglomerate surface exposed to the bonding and the difference between the maximum range of H-bond interaction and minimum interparticle distance (kg s ⁻²) (Chapter 5)
	Empirical constant in a correlation to predict U_{mf} (-) (Chapter 7)
ΔP	Differential pressure across two pressure taps in a CFB riser (kPa)
$\Delta P/\Delta h$	Change in pressure with respect to change in height in a CFB riser (kPa m ⁻¹)
ε_b	Volume fraction of the bubble phase i.e. the bubble phase holdup (-)
ε_{mf}	Bed voidage at minimum fluidization (-)
θ	Angle of the segment at which a sphere is cut to model a non-spherical particle (rad)
μ	Viscosity of gas mixture (kg m ⁻¹ s ⁻¹)
μ_f	Fluid viscosity (kg m ⁻¹ s ⁻¹)
μ_i, μ_j	Viscosity of species i and j , respectively (kg m ⁻¹ s ⁻¹)
ρ	Density of gas mixture (kg m ⁻³) (Chapter 11)
ρ^*	Density of primary nanoparticle agglomerate (kg m ⁻³)
ρ^{**}	Density of complex nanoparticle agglomerate (kg m ⁻³)
ρ, ρ_g	Fluid density (kg m ⁻³)
ρ_i	Density of component i (kg m ⁻³)
ρ_p	Particle density (kg m ⁻³)
$\rho_{p,mix}$	Characteristic particle density of a binary mixture (kg m ⁻³)
ρ_{p1}, ρ_{p2}	Particle densities of the two constituent species of a binary mixture (kg m ⁻³)
σ	Standard deviation of particle diameter of a mass-based particle size distribution (μm)

τ	Particle aspect ratio (-)
v_i, v_j	Atomic diffusion volumes of species i and j , respectively ($\text{m}^3 \text{mol}^{-1}$)
ϕ	Particle sphericity (-)
ϕ	Average cross-sectional solids holdup (-)
ϕ_1, ϕ_2	Particle sphericities of the two constituent species of a binary mixture (-)
ϕ_{mix}	Characteristic particle sphericity of a binary mixture (-)

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1

Introduction

1.1 Background

Gas-solid flow operations are integral to the chemical and processing industries. Some of the numerous applications involving gas-solid flows are solid-fuel combustion (e.g. pulverized coal combustion, solid waste incineration), catalytic cracking, mineral powder processing, chemical synthesis (e.g. Fischer-Tropsch synthesis), gas-solid separations (e.g. cyclones, electrostatic separators), chemical vapor deposition, heat transfer applications (e.g. nuclear reactor cooling) (Fan and Zhu, 1998c) etc. Natural phenomena involving gas-solid flow are moving of sand dunes, wind erosion, cosmic dust etc. Hence, physical understanding of gas-solid phenomena is crucial. Two of the most prevalent gas-solid flow operations are horizontal pneumatic conveying and fluidization.

1.1.1 Horizontal pneumatic conveying

Horizontal pneumatic conveying involves gas-assisted transport of solids through horizontal pipes. It offers various advantages like operational simplicity, flexibility, safety and low capital, operational and maintenance costs. Pneumatic conveying is employed in various industries like pharmaceutical, oil and gas, and agro-processing to transport materials like flour, granular chemicals, lime, soda ash, plastic chips, coal, gunpowder pellets, ores, grains etc. (Fan and Zhu, 1998b).

Knowledge of minimum conveying velocities is essential to ensure efficient operation of pneumatic conveying units. Three primary ‘critical’ velocities are usually defined in horizontal pneumatic conveying:

- Minimum saltation velocity (U_{salt}): The gas velocity in a horizontal line at which the particles start to drop out of suspension and settle on the bottom of the line (Cabrejos and Klinzing, 1994).
- Incipient motion velocity (U_{im}): The minimum gas velocity required to move a single particle initially at rest on the bottom surface of the pipe (Rabinovich and Kalman, 2009).
- Minimum pickup velocity (U_{pu}): The minimum gas velocity required to initiate motion of a particle initially at rest (Zenz, 1964).

Though ‘incipient motion’ and ‘pickup’ have been used interchangeably (Cabrejos and Klinzing, 1992, 1994; Hayden et al., 2003), U_{im} refers to the incipient motion of a single particle at rest on a surface (Rabinovich and Kalman, 2009), whereas, U_{pu} refers to the pickup of a particles from a layer or heap of particles (Cabrejos and Klinzing, 1992, 1994; Gomes and Mesquita, 2013; Hayden et al., 2003; Kalman et al., 2005). A point worth noting here is that the word ‘incipient’ defines the stage or phase of the operation of pneumatic conveying, i.e. pneumatic conveying has just begun and is in the early stage. U_{pu} and U_{salt} differ with respect to the initial state of a particle (Kalman et al., 2005). While U_{pu} refers to the minimum fluid velocity required to initiate motion in a particle from a layer of particles (Hallow, 1973), U_{salt} is associated with particles which are initially suspended in air and are being conveyed, and the critical velocity below which particles will be dropped out of flow and deposited at the bottom of the pipeline (Cabrejos and Klinzing, 1992). One defines the transition from a static to a dynamic state while the other defines a transition from the dynamic to the static state. Thus, the three critical velocities (namely, U_{salt} , U_{im} and U_{pu}) refer to distinctly different behaviors and/or environments. With respect to magnitude, $U_{pu} > U_{im} > U_{salt}$ (Rabinovich and Kalman, 2009).

Regarding applicability, U_{im} is almost exclusively used in theoretical analyses, since single individual particles are seldom encountered in horizontal pneumatic conveying. Out of U_{pu} and U_{salt} , the latter is more commonly used. However, U_{pu} is very crucial in certain applications and situations as listed below:

- Dilute flows, whereby a velocity higher than saltation velocity is required.
- If particles settle at the pipe bottom in cases of process upsets.
- Hydraulic conveying (Rabinovich and Kalman, 2008)
- Analyzing the movement of sand dunes and soil deposition in river and ocean (Kalman et al., 2005).
- Understanding erosion of silt on river beds (Rabinovich and Kalman, 2007) and wind erosion.
- Dust control applications (Gomes and Mesquita, 2013).
- Inhaler in drug delivery (Gandhimathi et al., 2015; Sham et al., 2004).

1.1.2 Fluidization

Fluidization is analogous to horizontal pneumatic conveying in the vertical gas-solid flow realm. It involves passing gas through a bed of solid particles thereby suspending the particles in a fluid-like state. Fluidized beds offer various well-acknowledged advantages like excellent inter-phase heat and mass transfer, high levels of intermixing of the particulate phase, uniformity in conditions, simplicity in geometric configuration, capability of continuous operation, applicability over a wide range of particle properties, suitability for large-scale operations and ease of operation due to the liquid-like flow of particles. The fluidic behavior allows the particles to be transported like a fluid leading to wide-ranging applications in industry, spanning coal gasification, energy production, fluid catalytic cracking, drying, calcining, production of polyethylene, biomass gasification and pharmaceuticals. Fig. 1.1

shows a schematic of a typical fluidized bed. The primary parts of a fluidized bed are the gas inlet, the plenum (which allows for distribution of gas before entering the bed), the distributor (which holds the bed and distributes gas evenly throughout the bed) and the particle bed (which is the fluidizing material). A fluidized bed may also contain a cyclone (used for separating gas from solids), a heat exchanger, an expanded section (which allows for reducing superficial gas velocity to reduce entrainment) and baffles.

Fig. 1.1. Schematic of a fluidized bed

The minimum fluidization velocity (U_{mf}), which is defined as the superficial gas velocity at which the drag force of the upward moving gas is equal to the apparent weight of the particle bed (Fan and Zhu, 1998b; Kunii and Levenspiel, 1991a), is a crucial parameter in

fluidization. Experimentally, U_{mf} is the superficial gas velocity at which the pressure drop across the particle bed plateaus or the expanded bed height starts increasing significantly. U_{pu} for pneumatic conveying is analogous to U_{mf} in fluidization systems (Babu et al., 1978; Bourgeois and Grenier, 1968; Ergun, 1952; Saxena and Vogel, 1977; Thonglimp et al., 1984; Wen and Yu, 1966) in that both dictate the minimum gas velocity required and have important implications in gas-solid flows, but the former is not as well-understood as the latter.

As the superficial gas velocity is increased above U_{mf} , the fluidized bed undergoes various regime changes like bubbling bed, turbulent bed, fast fluidization and dilute transport. Notably, for low gas velocities, bubbling fluidized beds are employed, which consist of a single column open to the atmosphere; and for high gas velocities Circulating fluidized beds (CFBs) are employed. CFBs are looped fluidized bed systems where the fluidizing particles are recycled back to the bed via a supporting system of pipes, cyclones and valves. CFBs find application, most notably in fluid catalytic cracking (FCC), chemical synthesis, coal gasification and more recently chemical looping due to several significant advantages like excellent heat and mass transfer, and catalyst or solids mobility.

1.1.3 Particle classification

The Geldart classification for powders undergoing fluidization was developed empirically by Geldart (1973). This is to date the most widely used classification for fluidizing particles. It is based on a plot of the difference between the densities of the fluidizing gas and bed particles ($\rho_p - \rho$) on the y-axis and the particle diameter (d_p) on the x-axis. Fig. 1.2 shows a simplified Geldart classification chart. The Geldart classification divides fluidizing powders into four categories (namely, C, A, B and D) depending on their fluidization behavior. Thus, this classification can be used to predict the fluidizing properties of a powder whose ρ_p and d_p values are known. Since the classification has been developed for room temperature and pressure and air as the fluidizing gas, some caution must be exercised while categorizing

powders undergoing fluidization at higher temperatures and pressures and with a different fluidizing gas than air. The four Geldart Groups are briefly described as follows:

- Geldart Group C: These are very fine and very cohesive particles with the inter-particle contact forces dominating. They are very difficult to fluidize and undergo gas-channeling or form plugs. Flour and cement are most common examples.
- Geldart Group A: These are fine and somewhat cohesive particles with inter-particle cohesion forces dominating, but hydrodynamic forces are also important. They undergo particulate fluidization and a maximum stable bubble size exists. Fluid Cracking Catalyst (FCC) particles is the most popular example.
- Geldart Group B: These are coarse non-cohesive particles with hydrodynamic forces dominating. They undergo bubbling fluidization and the bubble size increases with bed height. Building sand is a common example.
- Geldart Group D: These are very coarse particles and cannot undergo fluidization. They exhibit spouting beds. Biomass, food grains and gravel are common examples.

Fig. 1.2. Geldart (1973) classification for fluidizing powders

Particles undergoing pickup in horizontal pneumatic conveying are classified into three Pickup Zones (namely, I, II and III) according to their different pickup characteristics. These zones have a close correspondence to the four Groups of particles defined by Geldart (1973) (A, B, C and D). Specifically, Zone I corresponds to the coarse non-cohesive particles of Geldart Groups B and D, Zone II corresponds to the fine, somewhat cohesive Geldart Group A particles and Zone III corresponds to the very fine, cohesive Geldart Group C particles. A detailed description of the pickup Zones is given in Chapter 2.

1.2 Motivation and objectives

1.2.1 Overall objective

Despite being employed in the industry for decades, the analogous operations of horizontal pneumatic conveying and fluidization are still run based on experience and rules-of-thumb rather than physical models based on scientific principles. This may lead to decreased operational efficiency and wastage of resources, increased running and maintenance costs and reduced equipment life. Thus, this research effort endeavors to contribute to the physical understanding of these processes, specifically by assessing the effects of particle properties and operating conditions.

There are two common threads running through horizontal pneumatic conveying and fluidization. Firstly, both phenomena involve a physical interaction between the continuous gas phase and the discrete solid phase. Forces like the drag force of the gas, buoyancy, gravitational acceleration, friction and interparticle cohesion play important roles in this interaction. Secondly, particles possessing specific properties display distinct behaviors from others as seen from the Geldart Group and Pickup Zone classifications. But the close correspondence of these classifications shows that similar solids behave similarly across gas-solid flow operations. Also, despite particles in a Geldart Group (or impliedly Pickup Zone) displaying similar behavior, differing particle properties (Chew et al., 2011b; Gomes and Mesquita, 2013) or size distributions (Chew et al., 2011a; Chew et al., 2010) have been shown to cause behavioral changes even within the same Geldart Group (or impliedly Pickup Zone). Though there have been many studies (Chapters 2 and 8) documenting behavioral changes of different particles across and from Geldart Groups (and impliedly Pickup Zones), knowledge gaps (elaborated in the subsequent sections) still remain (e.g. the effect of polydispersity on U_{pu} , annulus flow behavior of Geldart Group B particles). Thus, the aim here is to identify and

investigate such gaps in horizontal pneumatic conveying and fluidization, hence aiding the progress towards first principles models.

The tree in Fig. 1.3a shows the structure of the studies undertaken to further the physical understanding of gas-solid flow. Horizontal pneumatic conveying falls under horizontal flow, where four studies were conducted on the minimum pickup velocity (U_{pu}), namely, two on the U_{pu} of micron-scale particles (Geldart Groups B and D), one on that of nano-scale particles (Geldart Group C) and one bridging the gap between micron-scale and nano-scale particles (Geldart Groups C and A). Three studies were conducted under vertical flow on fluidization, namely, a review of correlations predicting the minimum fluidization velocity, U_{mf} (Geldart Groups A, B and D); and two studies on circulating fluidized beds (CFBs) assessing pressure signals and the annulus flow behavior of Geldart Group B particles. Reiterating, the common thread was the different particle behaviors across or within Geldart Groups (or impliedly Pickup Zones).

Fluidized beds are ultimately employed to carry out chemical reactions. Hence, along with a physical understanding of the gas-solid interaction, examining the behavior of chemical reactions in fluidized beds is important. The modeling study, represented in Fig. 1.3b, thus examines the effects of particle properties and operating conditions on methane (CH_4) production via a carbon-dioxide (CO_2) methanation reaction in a bubbling fluidized bed.

(a) Physical understanding **(b) Reactor modeling** **Fig. 1.3.** Structure tree: (a) Physical understanding and (b) Reactor modeling**1.2.2 Horizontal pneumatic conveying**

As elucidated above, minimum pickup velocity (U_{pu}) is a very essential minimum conveying velocity. Having a mechanistic understanding of U_{pu} will allow for more efficient processes to be designed and operated. In addition, attrition and fragmentation of particles is a significant problem in industrial pneumatic conveying lines. Hence, if the transport velocity is kept minimum based on knowledge of U_{pu} , particle breakage can be reduced considerably in dilute phase transport systems (Kalman, 2000; Salman et al., 2002). Despite the advances on U_{pu} to date, Gomes and Mesquita (2014) very recently compared various U_{pu} correlations and detected discrepancies of about an order-of-magnitude in U_{pu} predictions and contradictory

shapes (convex/ concave) for the U_{pu} versus particle diameter (d_p) trends, which signals the need for improved mechanistic understanding. Although in practice particle systems are typically polydisperse, much of the earlier work has focused on monodisperse systems or only reported d_p ranges without specifying polydispersity. Particle geometry is another important factor, which affects particle flow behavior in a gas via the drag force and the distribution of boundary layer on the surface (Fan and Zhu, 1998a). Particle shape thus affects the flowability of particle powders and also their packing. Most analyses in fluid dynamics in general, and studies on U_{pu} in particular, are done on spherical particles. Particle sphericity (ϕ) best describes the magnitude of departure of an irregular particle from a perfect sphere, and hence is critical to the description of the behavior of non-spherical particles. Information on the impact of ϕ on U_{pu} is scarce. Hence, current experimental efforts aim at revealing the impact of two prevalent and important parameters on U_{pu} , namely (a) the width of continuous (lognormal) particle size distribution (PSD) and (b) particle sphericity (ϕ); and investigating binary mixtures with constituents differing in particle diameter (d_p), particle density (ρ_p) and particle sphericity (ϕ) with respect to U_{pu} .

Previous studies on U_{pu} have focused on micron-sized particles in the size range of 40 – 3000 μm (Cabrejos and Klinzing, 1992; Kalman et al., 2005; Rabinovich and Kalman, 2008). However, no understanding on the U_{pu} of nanoparticles is available to date, even though the processing of nanoparticles is becoming increasingly important (van Ommen et al., 2015). Such knowledge is not only important for industrial processing of nanoparticles, but also to increase insight in nanoparticle dispersion in workplaces or the environment (Kuhlbusch et al., 2011; Kumar et al., 2011). Regarding gas–solid flow of nanoparticles, considerable research has been carried out on fluidization, which has been harnessed for coating nanoparticles by means of atomic layer deposition (ALD) (Goulas and van Ommen, 2014; King et al., 2012). Even so, understanding on the pneumatic conveying of nanoparticles is lacking, despite the importance

spanning pneumatic dispersion in nanoparticle synthesis (Shabeer et al., 2007), nanoparticle inhaler in drug delivery (Gandhimathi et al., 2015; Sham et al., 2004), and environmental pollution by nanoparticles (Niu et al., 2010; Stahlmecke et al., 2009). Moreover, empirical or semi-empirical models (Cabrejos and Klinzing, 1994; Kalman et al., 2005) pertaining to U_{pu} developed so far are based on micro-scale particles only. Hence, the first experimental study on nanoparticles is aimed at determining the U_{pu} values for nanoparticles and differentiating between behaviors of polar and apolar nanoparticles with respect to U_{pu} . Based on the results of the first study (Anantharaman et al., 2015), which revealed that the behavior of nano-scale particles cannot be predicted by the classification developed based on micro-scale particles, the second study on U_{pu} of nanoparticles aims at bridging the gap in terms of (i) understanding the underlying discrepancy in behavior between the nano-scale particles and the lower range of micro-scale particles (i.e., Zone III or Geldart Group C), and (ii) determining when the transition between nano-scale and micro-scale occurs.

1.2.3 Fluidization

After the first commercially viable fluidized bed, the Winkler coal gasifier, was implemented more than seven decades ago (Kunii and Levenspiel, 1991b), numerous correlations on various aspects of the fluidization behavior have been developed towards a more mechanistic understanding and a better predictive capability. Unfortunately, instead of advancing the knowledge base, the correlations available for entrainment (Chew et al., 2015) and transport disengaging height (TDH) (Cahyadi et al., 2015) have been found to give predictions that are orders-of-magnitude apart, which limits the applicability and hampers understanding. The extensive discrepancies for the different entrainment and TDH correlations are stemmed in the more complex behaviors (e.g., origins of entrainment (Werther and Hartge, 2003), clustering (Cahyadi et al., 2017)) associated with such higher-velocity fluidization regimes, which hence leads to results varying significantly for the different reactors and/or

instruments used for collecting the experimental data used in developing the best-fitted empirical correlation. Comparatively, the U_{mf} seems to be a more tractable parameter in fluidization. The experimental measurement is simpler and less subject to variations due to differences of reactors or instruments or analysis methods. However, more than a hundred correlations have sprouted since 1950 to enable the prediction of the U_{mf} value. Therefore, the overall goal of the first fluidization study is to review the minimum fluidization velocity (U_{mf}) correlations available in literature in order to shed light on the state of such understandings. It attempts to advance the understanding or lack thereof of U_{mf} through a comparative analysis of the correlations available in literature. The accurate knowledge of U_{mf} is vital for the design and development of fluidized beds (Hartman and Coughlin, 1993; Lippens and Mulder, 1993). U_{mf} is similarly important for research on segregation, the separation or de-mixing of different sized particles in the column due to fluidization (Cui and Grace, 2007; Rowe and Nienow, 1976; Tang and Puri, 2004), in fluidized bed systems. Operating superficial gas velocities in studies on segregation (Cahyadi et al., 2017; Cahyadi et al., 2015; Chew and Hrenya, 2011; Chew et al., 2010; Curtis and Van Wachem, 2004; Formisani, 1991; Gauthier, 2009; Horio, 2010; Kunii and Levenspiel, 1991b; Muzzio et al., 2002; Royer et al., 2009; Sundaresan, 2001) are decided on the basis of U_{mf} values. Therefore, a comprehensive analysis of U_{mf} correlations will be an advantageous undertaking.

Among fluidized bed systems, Circulating Fluidized Beds (CFBs) are utilized in a wide range of industrial processes as mentioned earlier. However, despite the widespread operation of such systems, some challenges remain like undesirable flow behavior, back-mixing, erosion, particle attrition and flow instabilities during operations (Morrow, 1985, 1988). The lack of tools to trouble-shoot and optimize such processes is a key factor in addressing these challenges. The primary section of a CFB is the riser, which is the vertical sector where the gas-solid mixture flows with high velocity. Variations and fluctuations in the solids velocity

and concentration in the riser are not infrequent. This results in non-uniform gas-solid contact in the riser across space and time, which affects the efficiency of the whole unit. Most studies documenting these changes have been published for Geldart (Geldart, 1973) Group A particles, where cluster instabilities and the core-annulus flow structure (a relatively dilute core and a dense annulus) have been well-recorded (Fan and Zhu, 1998d; Kunii and Levenspiel, 1991c). Geldart Group B particles are well-acknowledged to fluidize differently from Geldart Group A particles, with the most significant difference being the lack of cohesion in the former (Geldart, 1973). Because behavioral trends of Geldart Group A particles cannot be directly extrapolated to predict those of Group B, a separate investigation into the hydrodynamics of the latter is warranted. Though fluidization of the larger and denser Geldart Group B particles will require more energy than that of Geldart Group A particles, they are gaining increasing prominence in (i) CFB combustors (Berruti et al., 1995; Bi and Liu, 2010; Mahmoudi et al., 2012), (ii) CFB gasifiers of coal and biomass (Bi and Liu, 2010; Chang et al., 2016; Mahmoudi et al., 2012), (iii) biomass pyrolysis in CFBs as inert materials to improve the fluidization quality of biomass (Boukis et al., 2007; Cui and Grace, 2007; Dai et al., 2001; Mahmoudi et al., 2012; Oliveira et al., 2013; Paudel and Feng, 2013) and (iv) chemical looping combustion (CLC) in CFB reactors (Li et al., 2011; Rodrigues et al., 2015). Regarding (iv), Geldart Group B particles allow the use of non-mechanical valves required to be used at high temperatures (~1173 K) needed for CLC (Yazdanpanah et al., 2012) and can also be easily separated from the unburned solid fuel at the reactor exit (Rodrigues et al., 2015). Previous studies on Geldart Group B particles have investigated cluster characteristics (Chew et al., 2012a, b), mass flux (Chew et al., 2011b), species segregation (Chew et al., 2011a), radial solids concentration (Chew et al., 2012c). The two studies in this thesis look into the energy of pressure signals and upflow annulus behavior of Geldart Group B particles in a CFB riser. The goal is to contribute in establishing a rich dataset for Geldart Group B riser characteristics.

Pressure transmitters are the most common instrumentation in commercial fluidized beds, due to the ease of measurement even under harsh conditions, and also to their robustness, low-cost and non-intrusive nature (which allows for measurements to be taken without distorting the flow) (Tahmasebpour et al., 2013; van Ommen et al., 2011). Another instrumentation available for routine measurement is for temperature (Werther, 1999). However, the temperature signal features a dynamic much slower than the pressure signal dynamic, since the latter is based on pneumatics. Hence, the data obtained from pressure transmitters are often the only useful data available for understanding the hydrodynamics in fluidized beds. This necessitates efforts to extract as much valuable information from pressure signals as possible. Hence, differential pressure (ΔP) signals from a CFB riser of Geldart Group B particles are assessed with a view to obtain (i) a mechanistic understanding on the hydrodynamics (e.g., gas-particle interaction), and (ii) more in-depth information regarding the process. The objective is to evaluate the effectiveness of the ubiquitous ΔP signals in differentiating operating conditions and material property.

The core-annulus phenomenon in a CFB riser of Geldart Group A particles was first proposed by Bierl and Gajdos in 1982 (Bierl and Gajdos, 1982). They proposed a flow structure, which consisted of a dilute core of upward moving particles, which occupied most of the cross-section, and a relatively dense annulus at the riser wall, which consisted mainly of downward-moving solids. A typical core-annulus structure is depicted in Fig. 1.4.

Fig. 1.4. Schematic of the core-annulus flow structure in a CFB riser

Although there has been some research on demarcating flow regimes in CFB risers containing Geldart Group B particles (Das et al., 2008), none has focused on investigating the core-annulus behavior of these systems. A previous study has shown surprisingly that Geldart Group B particles exhibited a reverse core-annulus behavior (i.e., a dense core and a dilute annulus), due to the higher Stokes numbers (St) of the particles (Chew et al., 2012c). The lack of knowledge on the impact of polydispersity, which is an ever-present aspect of solids processing in industry, has been flagged as being a critical gap to bridge (Curtis and Van Wachem, 2004; Hrenya, 2010; Muzzio et al., 2002; Sundaresan, 2001). Polydisperse systems have been shown to exhibit distinctly different behaviors from monodisperse systems (Chew et al., 2011a; Fan et al., 1990; Hrenya, 2010; Ottino and Khakhar, 2000; Rowe and Nienow, 1976; van Ommen et al., 2011). Distinctly different behaviors among monodisperse systems and different types of polydispersity (e.g., binary mixtures and continuous particle size distributions (PSDs)) have been reported in bubbling fluidized beds (Chew and Hrenya, 2011; Chew et al., 2010; Hoffmann and Romp, 1991; Lin et al., 2002; van Ommen et al., 2011) and circulating fluidized beds (CFBs) (Cahyadi et al., 2015; Chew et al., 2015; Chew et al., 2013; Issangya et al., 2010).

The impact of polydispersity on the annulus flow behavior has, however, not been studied. Accordingly, the second CFB riser study is focused on understanding the annulus flow behavior of Geldart Group B particles and how polydispersity impacts it.

1.2.4 Reactor modeling

In addition to attempt at contributing to the mechanistic understanding of fluidization systems, an effort is made here to investigate the effects of fluidized bed hydrodynamics and operating conditions on reactor performance.

The methanation of carbon-dioxide (CO_2) to convert it into Synthetic Natural Gas (SNG), i.e. methane, is an effective way (Centi and Perathoner, 2009; Centi et al., 2013; Hoekman et al., 2010; Miguel et al., 2015) of tackling both problems of effective CO_2 capture on one hand and finding a renewable source of energy on the other. Anthropogenic emissions due to rapid burning of fossil fuels are the primary contributors to atmospheric CO_2 , which is well-acknowledged to cause global warming (Zhang et al., 2014). Today, the major share of industrially used methane comes from fossil fuels, which are fast depleting (Rönsch et al., 2016). Electricity storage is gaining prominence as an environmentally sustainable application of CO_2 methanation. This is associated with the global CO_2 recycling proposed by Hashimoto et al. (1999), which will cause an estimated 79% reduction in CO_2 emissions. The process involves electrolysis of seawater with electricity obtained via desert solar panels to produce hydrogen, H_2 , which in turn is used for the CO_2 methanation reaction. The methane thus obtained is then supplied to power plants, which then supply back the CO_2 produced there. Hence, though the CO_2 methanation reaction requires elevated temperatures ($> 520 \text{ K}$), associated energy costs would far outweigh the benefits when integrated with electrolysis in a CO_2 recycling system (Hashimoto et al., 1999). Hence, research on CO_2 methanation has regained importance in recent times. However, though known for a very long time, a standard commercial process has not yet been established for the CO_2 methanation (Sabatier) reaction

due to issues associated with kinetics, effective catalysts and reactor designing (Schlereth and Hinrichsen, 2014).

All modelling studies on CO₂ methanation have been carried out on fixed-bed reactors. However, fluidized bed reactors offer various well-acknowledged advantages like excellent inter-phase heat and mass transfer, and ease of operation, among others. There have been a few studies (Kopyscinski et al., 2011a, b; Zhang et al., 2015) on CO methanation involving fluidized beds but none on CO₂. Also, a majority of the reactor-modelling studies on the Sabatier reaction focus on its heat removal aspects. But as Kopyscinski et al. (2010) and Chein et al. (2016) have observed, the CO₂ methanation reaction is cognizably sensitive to parameters like temperature, pressure, inlet feed composition etc. Also, reactor studies (Kopyscinski et al., 2011a, b; Zhang et al., 2015) on CO methanation in fluidized beds have highlighted the importance of hydrodynamics in product yield. Hence, the current study attempts to investigate the effects of temperature, pressure, inlet feed ratio, inlet superficial gas velocity, particle diameter, particle density, particle sphericity and the bed voidage at minimum fluidization on methane production by the Sabatier reaction through a heterogeneous bubbling fluidized bed reactor model.

1.3 Structure of the thesis

This thesis is divided into three parts focusing on pneumatic conveying, fluidization and reactor modelling respectively. Part I (Chapters 2-6) deals with the minimum pickup velocity (U_{pu}), Part II (Chapters 7-10) discusses the minimum fluidization velocity (U_{mf}), and differential pressure signals and radial mass flux distribution in CFB risers and Part III (Chapter 11) attempts numerical modeling of a carbon-dioxide (CO₂) methanation bubbling fluidized bed reactor.

Chapter 1 provides the reader a background of the crucial gas-solid flow operations of horizontal pneumatic conveying and fluidization, gives the motivation behind this research effort by elucidating current knowledge gaps and the research objectives that were planned to be achieved.

Chapter 2 provides a comprehensive literature survey of the research works associated with U_{pu} .

Chapter 3 deals with the effects of continuous particle size distribution (PSD) width and particle sphericity on U_{pu} . Experiments on three different materials and five different PSD widths reveal the discrepancy between the U_{pu} values of the experimental data here and values predicted by available correlations underscores the non-negligible impact of PSD width and particle sphericity.

Chapter 4 discusses the impacts of particle diameter (d_p), particle density (ρ_p) and particle sphericity (ϕ) on the minimum pickup velocity (U_{pu}) of binary mixtures of Geldart group B particles. Binary-size (constituents vary only in d_p), binary-density (constituents vary only in ρ_p) and binary-size-density (constituents vary in d_p and ρ_p) mixtures are considered. A force balance analysis incorporating ϕ is also conducted.

Chapter 5 reports the U_{pu} values of nanoparticles. Six nanoparticle species of different materials and surfaces (polar and apolar) were considered. The differences between behaviors of nano-scale and micro-scale particles and polar and apolar nanoparticles in terms of U_{pu} were noted. U_{pu} values of nanoparticles were compared against predictions of the Three-Zone model developed for micro-scale particles.

Chapter 6 attempts to reveal the missing link between nano-scale and micro-scale particles in terms of U_{pu} . Experiments were conducted on nine alumina particles with particle diameters (d_p) ranging from 5 nm to 110,000 nm to determine their U_{pu} , which was then

compared against the established pickup Zones. Modifications to the Three-Zone model are also proposed.

Chapter 7 attempts to provide a comprehensive comparison of the U_{mf} values predicted by over a hundred correlations available in literature, which are classified into four types depending on the form of the equation and applied to more popular Geldart Groups A, B and D particles. Also, a comparison of the correlations containing either voidage at minimum fluidization (ε_{mf}) and/or particle sphericity (ϕ) is carried out.

Chapter 8 provides a comprehensive literature survey of the research works on CFBs particularly in context of pressure signal analysis and the core-annulus structure.

Chapter 9 is targeted at assessing the hydrodynamic characteristics that can be understood from a comprehensive dataset of pressure signals via wavelet decomposition. The effects of material property, operating condition and riser axial position on the differential pressure (ΔP) signals were evaluated for five Geldart Group B particle systems by comparing the energy contained in the wavelet-decomposed signals (E_{Dj}).

Chapter 10 studies the annulus flow behavior of Geldart Group B particles in a CFB riser. The effects of material property, operating conditions and polydispersity were revealed by studying for three monodisperse particles, two binary mixtures and one continuous particle size distribution. The data were compared to predictions available from flow regime maps.

Chapter 11 endeavors to apply a two-phase heterogeneous model to a carbon-dioxide (CO_2) methanation bubbling fluidized bed reactor and assess the effects of varying operating conditions and bed properties on the generation of synthetic natural gas, methane (CH_4).

Chapter 12 gives brief takeaways from each of the studies presented in this thesis and an overall conclusion, and provides perspectives on potential future work.

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Part I

Horizontal Pneumatic Conveying

2

Literature review – minimum pickup velocity

Studies on pneumatic conveying systems have been conducted on various aspects such as characterization of pneumatic flow into various flow modes and how properties of materials affect it (Pan, 1999), studies on swirling in axial and radial directions (Li and Tomita, 2000), limitations on transport in pneumatic conveying (Wypych and Yi, 2003) simulations which consider shear rate (Stevenson et al., 2002), and a comparison of various models used to simulate pneumatic flow including Eulerian-Eulerian, Eulerian-Lagrangian and hybrid approaches on the basis of their ability to predict system behavior during pneumatic conveying (Pirker et al., 2010). The various ‘critical’ velocities associated with horizontal pneumatic conveying have been discussed in Section 1.1.1. Recall that minimum pickup velocity (U_{pu}) is defined as the gas velocity required to initiate the rolling motion of or suspend the particle initially at rest (Hallow, 1973; Kalman et al., 2005). The word ‘pickup’ is associated with the gas velocity which is responsible for initiating pickup or taking the stationary system into the incipient stage of pickup.

One of the early works on U_{pu} was by Cabrejos and Klinzing (1992) who proposed a technique based on visual observation to determine U_{pu} . Particles of diameter (d_p) between 10 and 800 μm were investigated and a semi-empirical correlation involving Reynolds (Re) and Archimedes (Ar) numbers was developed. Notably, U_{pu} was shown to first increase and then decrease with increasing d_p . This was attributed to decreasing inter-particle cohesion in the lower d_p range, and increasing inertial forces in the higher d_p range. Using dimensional analysis, Cabrejos and Klinzing (1994b) developed a correlation for U_{pu} as a function of particle size

(d_p), particle Reynolds number (Re_p), particle density (ρ_p) and gas density (ρ) using dimensional analysis in 1994:

$$\frac{U_{pu}}{\sqrt{gd_p}} = 0.0428 Re_p^{0.175} \left(\frac{D}{d_p} \right)^{0.25} \left(\frac{\rho_p}{\rho} \right)^{0.75} \quad (2.1)$$

About a decade later, (Hayden et al., 2003) investigated monodisperse Geldart Groups A and C (Geldart, 1973) particles, and observed (i) similarly a minimum in the plot of U_{pu} versus d_p , depending on the relative dominance of cohesive and inertial forces, (ii) that electrostatic forces play an important role for smaller particles (with diameters of 20-40 μm), resulting in an increase in U_{pu} , particularly for the insulating glass particles rather than the conducting stainless steel particles, and (iii) that non-spherical particles in general have greater U_{pu} values than spherical particles, although detailed investigation on the effect of particle sphericity was not reported.

Kalman et al. (2005) after conducting experiments on particles from Geldart groups A, B and D, compiled their results with ~60 datapoints found in literature (Cabrejos and Klinzing, 1994a; Cabrejos and Klinzing, 1992, 1994b; Hayden et al., 2003; Ramadan et al., 2003) and defined three Pickup Zones (I, II and III) for classifying particles according to their different pickup characteristics. These zones have a close correspondence to the four Groups of particles defined by Geldart (1973) (A, B, C and D) according to their fluidization characteristics as observed in bubbling fluidized beds. The Zones were classified based on relationships between the Archimedes number, (Ar), and the modified particle Reynolds number (Re_p^*). Both, the Reynolds and Archimedes numbers are dimensionless groups with the former being the ratio between inertial and viscous forces and the latter, the ratio between gravitational and viscous forces. The resulting curve of particle Re_p^* versus Ar has been termed as the master curve for minimum pickup velocity (Rabinovich and Kalman, 2008). Particles have been classified into three Zones based on the following equations (Kalman et al., 2005):

$$\text{Zone I: } \text{Re}_p^* = 5\text{Ar}^{\frac{3}{7}} \text{ for } \text{Ar} \geq 16.5 \quad (2.2)$$

$$\text{Zone II: } \text{Re}_p^* = 16.7 \text{ for } 0.45 < \text{Ar} < 16.5 \quad (2.3)$$

$$\text{Zone III: } \text{Re}_p^* = 21.8\text{Ar}^{\frac{1}{3}} \text{ for } \text{Ar} \leq 0.45 \quad (2.4)$$

The particle Reynolds number is modified to account for different pipe diameters in the following manner (Kalman et al., 2005):

$$\text{Re}_p^* = \frac{\rho d_p U_{pu}}{\mu_f \left(1.4 - 0.8e^{-\frac{D/D_{ref}}{1.5}} \right)} \quad (2.5)$$

The Archimedes number is defined as:

$$\text{Ar} = \frac{g\rho(\rho_p - \rho)d_p^3}{\mu_f^2} \quad (2.6)$$

where D_{ref} is the reference pipe diameter of 50 mm. Approximately, Zone I corresponds to Geldart Groups B and D particles which are coarser and dominated by inertial forces, Zone II corresponds Geldart Group A particles which are finer and dominated more by cohesive forces than inertial forces and Zone III corresponds to Geldart group C particles which are extremely fine and highly cohesive with inertial forces virtually absent.

Fig. 2.1 shows Re_p^* vs Ar according to the Three-Zone Model (Kalman et al., 2005), while Fig. 2.2 shows U_{pu} vs d_p as calculated by the Three-Zone Model (Kalman et al., 2005) for glass beads ($\rho_p = 2500 \text{ kg/m}^3$). For Zone 1 (Geldart Groups B and D) particles, increasing d_p will increase inertial forces only, since cohesive forces are virtually absent in these type of particles. Hence, for Zone I particles, Re_p^* and U_{pu} increase as Ar and d_p increase, respectively. In Zone II (Geldart Group A) particles, inter-particle cohesive forces are more dominant than inertial forces and increase with decreasing d_p , thus increasing U_{pu} . Hence, for Zone II particles, U_{pu} is inversely proportional to d_p , thereby maintaining a constant value of Re_p^* with respect

to Ar. In Zone III (Geldart Group C), particles are so cohesive that they are picked up as clouds instead of individually. Hence, for Zone III particles, U_{pu} remains unaffected by change in d_p and Re_p^* decreases with decreasing Ar.

Fig. 2.1. The Three-Zone Model of (Kalman et al., 2005), developed from ~60 datapoints from five previous works (Cabrejos and Klinzing, 1994a; Cabrejos and Klinzing, 1992, 1994b; Hayden et al., 2003; Ramadan et al., 2003). The x -axis represents the Archimedes number (Ar) and the y -axis represents the modified particle Reynolds number (Re_p^*).

Fig. 2.2. U_{pu} vs d_p as calculated by the Three-Zone Model (Kalman et al., 2005) for glass beads ($\rho_p = 2500 \text{ kg/m}^3$)

Furthermore, Rabinovich and Kalman (2009) scrutinized the pickup phenomenon from particle deposits as opposed to pickup from a layer of particles, and noted that U_{pu} from deposits is strongly dependent on the height and shape of the particle heap.

Earlier, experiments on U_{pu} have largely focused on monodisperse particles, or rather has assumed monodispersity and/or have only reported particle size ranges but not the exact particle size distribution (PSD) investigated. However, practical operations are typically polydisperse. More recently, Goy et al. (2011) investigated the effects of binary mixtures of particles belonging to different zones on U_{pu} . For binary mixtures involving only Zone I particles, the minimum pickup velocity increases as the mass fraction of the larger particles increases thus mimicking the monodisperse trend. For binary mixtures of Zones I and II

particles, the U_{pu} decreases with an increase in the mass fraction of the larger particles, with the cohesive forces of the smaller Zone II particles becoming dominant; whereas for binary mixtures of Zones I and III particles the U_{pu} of Zone I remain unaffected in spite of the presence of Zone III particles showing that inertial forces of Zone I particles completely dominate cohesive forces of the Zone III particles. The equations defining particle Zones can only predict correctly the minimum pickup velocities of binary mixtures of only Zone I particles, and those constituting both Zone I and II particles, whereas they fail to do the same for binary mixtures constituting both Zone I and III particles. The trend of U_{pu} with respect to mass fraction depends on the ratio of diameters of the two species involved.

Furthermore, (Tay et al., 2012) investigated the effects of ternary mixtures (i.e., three particle species) on U_{pu} and hypothesized that, up to a limit of 20% by mass of the smallest particles, the smallest particles aid entrainment of the larger particles by occupying the interstitial spaces thereby enhancing the interstitial velocity. A new Re_p^* versus Ar master curve for binary and ternary mixtures was developed based on the Sauter's mean of the mixtures (Tay et al., 2012):

$$Re_p^* = 20Ar^{\frac{1}{4}} \text{ for } 20 \leq Ar_{\text{mixture}} \leq 2500 \quad (2.7)$$

There is scarce information on the impact of particle sphericity (ϕ), on U_{pu} . Hayden et al. (2003) presented one of the first efforts, which showed that non-spherical particles exhibited a greater U_{pu} than spherical particles. Kalman et al. (2005) measured U_{pu} of non-spherical Geldart group D particles and observed discrepancies between the measured U_{pu} values and those predicted by the Three-Zone model of Eqs. (2.2), (2.3) and (2.4). To account for the effect of particle sphericity (ϕ), they defined a characteristic Archimedes number (Ar) by modifying Eq. (2.6) and found that non-spherical particles displayed a lower characteristic Ar than spherical ones. Gomes and Mesquita (2013) took an analytical approach in developing a pickup

velocity model. Its utility is, however, limited beyond the specific system investigated, because we observed errors of more than 40% in the model prediction even with their own data, and they used empirical coefficients (e.g., fractional powers of U) to improve the model prediction. The same authors Gomes and Mesquita (2014) also recently reviewed all available U_{pu} correlations and found that their model predictions were contradictory.

All studies on U_{pu} only involve micron-sized particles. Knowledge of the U_{pu} of nanoparticles is absent. With the advent of nanotechnology in view of the benefits associated with the high surface area, the pneumatic conveying of nanoparticles warrants more understanding. Regarding gas-solid flow of nanoparticles, considerable research has been carried out on fluidization, which has been harnessed for coating nanoparticles by means of atomic layer deposition (ALD) (Goulas and van Ommen, 2014; King et al., 2012). Unsurprisingly, nanoparticles fluidize as agglomerates as opposed to individual particles (Nakamura and Watano, 2008; van Ommen et al., 2012). Multistage agglomerate fluidization (i.e., formation of nanoparticle agglomerates in stages during fluidization) has been reported, with primary agglomerates as the building blocks of complex agglomerates (Yao et al., 2002). Along with van der Waals, electrostatic and capillary forces of attraction in fluidizing nanoparticles, it has been shown that hydrogen bonds also play a non-negligible role in agglomerate formation and fluidization characteristics of nanoparticles (Tahmasebpour et al., 2013). Specifically, polar and apolar nanoparticles exhibit very different fluidization behaviors and minimum fluidization velocities (U_{mf}) (Tahmasebpour et al., 2013). De Martin and van Ommen (de Martín and van Ommen, 2013) developed a model to estimate the size and density of fluidized nanoparticle agglomerates. Various methods of observing and characterizing nanoparticle agglomerates have been developed (de Martín et al., 2013), and understanding on different fractal dimensions of nanoparticle agglomerates have also been reported, with the most common fractal dimension reported being 2.5 (Nam et al., 2004; Quintanilla et al., 2012;

Valverde and Castellanos, 2007, 2008). De Martin et al. (de Martin et al., 2014; de Martín et al., 2014) further revealed that each hierarchical stage of nanoparticle agglomerate formation has a different fractal dimension. Though considerable work has been carried out on the fluidization of nanoparticles, understanding on the pneumatic conveying of nanoparticles is lacking, despite the importance spanning pneumatic dispersion in nanoparticle synthesis (Shabeer et al., 2007), nanoparticle inhaler in drug delivery (Gandhimathi et al., 2015; Sham et al., 2004), and environmental pollution by nanoparticles (Niu et al., 2010; Stahlmecke et al., 2009).

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3

Effect of continuous particle size distribution (PSD) and particle shape on minimum pickup velocity

This chapter deals with the effects of continuous particle size distribution (PSD) width and particle sphericity on minimum pickup velocity (U_{pu}).

3.1 Experimental protocol

3.1.1 Experimental setup

Fig. 3.1 shows a schematic diagram of the setup used, which is identical to that in (Goy et al., 2011) similarly for determining minimum pickup velocity (U_{pu}). Specifically, the setup consists of three horizontal sections (namely, Sections A, B and C), connected by means of flanges, and made of acrylic with an internal diameter of 15.5 mm and wall thickness of 2 mm. With regards to the internal diameter, (Kalman et al., 2005) recognized the effect of pipe diameter on U_{pu} , and hence proposed a modified Reynolds number (Re_p^* , given in Eq. (2.5)) to allow for synchronization of U_{pu} values across measurements in different setups. Accordingly, we have used Re_p^* in our analysis with a wetted diameter of the semi-circular cross-section, 9.47 mm (Goy et al., 2011), to facilitate comparison with previous work.

Section A is the inlet for air, the supply of which comes from a central air compressor with a maximum flow rate of 0.001 m³/s. The length of Section A is designed to be 91 cm to ensure that the air velocity profile is fully-developed before entraining the particles contained in Section B. Section B is 30 cm long and divided into two semi-cylindrical halves along the horizontal axis; the bottom half is completely packed with plasticine for a length of 27.5 cm from Section A, while the remaining 2.5 cm provides a compartment in which the particles to

be investigated are loaded. It should be noted that air only flows through the top half semi-circular cross-sectional area in Section B. Section C is 83 cm long to minimize exit effects; the end is connected to a cyclone collector. The key feature of this setup is to maintain a constant cross-sectional area for air flow, which in turn maintains a constant value of air velocity (Goy et al., 2011; Tay et al., 2012). Specifically, the free cross-sectional area for air flow remains constant throughout the experiment irrespective of the extent of particle entrainment, as opposed to a setup whereby the particles are placed in a heap in the pipe (Cabrejos and Klinzing, 1992, 1994), which causes the cross-sectional area to increase as particle entrainment progresses.

Fig. 3.1. Experimental setup for measuring minimum pickup velocity (a) Various components (b) Setup in use

3.1.2 Materials

Geldart Group B (Geldart, 1973) particles of a wide size range, various particle sphericities (ϕ), and various particle densities (ρ_p) were used for the experiments. The reason for investigating Geldart Group B is to eliminate the effects of inter-particle cohesive effects, such that only the impacts of polydispersity and particle shape are elucidated. The three particle species used were glass, plastic and aluminum oxide, which were purchased from Abrasive Engineering Pte Ltd. (Singapore). Table 3.1 lists the relevant particle properties. The method used to determine ϕ is described in the next section.

Table 3.1. Relevant particle properties of the materials investigated

Material	ρ_p (kg/m³)	ϕ (-)
Glass	2500	0.97
Plastic	1150	0.74
Aluminum Oxide	3920	0.82

3.1.3 Determining particle sphericity

The three-dimensional sphericity of a particle is defined as the ratio of the surface area of a sphere with the same volume as the particle to the surface area of the particle (Kunii and Levenspiel, 1991a). However, these sphericity values are not readily available (refer Chapter 7) and the best method to determine them experimentally is only via data-fitting (Kunii and Levenspiel, 1991b). Hence, the sphericity of particles used in this study was determined on the basis of the projected image of the particle. Admittedly, such a definition would be biased as the particle would only rest on its most stable plane and the sphericity would be calculated based on the corresponding projected image. Nevertheless, the plane on which a particle rests is the important one as far as U_{pu} is concerned, since the drag force on the particle is determined based on the projected area of this plane as will be seen in the next chapter.

Various ways to define the shape factor of a particle based on its two-dimensional projected image are available in literature (Cavarretta et al., 2009; Doroodchi et al., 2013; Geldart, 1990; Waddell, 1933). In this work, the definition by Waddell (1933) based on projected area of the particle was considered. It should be noted that Waddell represents the first paper involving the determination of particle sphericity (ϕ) based on the projected area of the particle (A), and most subsequent papers are based on variations of the Waddell expression. Specifically, the definition of ϕ by Waddell (1933) is a function of A and the diameter of the minimum circumscribed circle around the particle (D_{cmin}):

$$\phi = \frac{2\sqrt{A/\pi}}{D_{cmin}} \quad (3.1)$$

Fig. 3.2. Microscope images of (a) glass, (b) aluminum oxide and (c) plastic particles

In this work, the ϕ values of the particles were determined via microscopic imaging. The images of the particles were obtained with a microscope (Olympus BX51), then the outlines of the two-dimensional projections extracted using AutoCad 2010 in order to apply Eq. (3.1). Fig. 3.2 shows that the three species, namely, glass, aluminum oxide and plastic particles, clearly have different particle sphericities. For each material, images of up to 15 particles of various sizes were obtained and the ϕ values averaged. The ϕ values of the three materials so calculated are displayed in Table 3.1 and Fig. 3.3, where the error bars represent two standard deviations. Specifically, the standard deviation of each set of ϕ values obtained from different particles for a material was calculated. This value was then added to and subtracted from the average ϕ value to obtain respectively, the lower and upper bounds of the

error bars. Statistically, ϕ values of about 70% of all particles of each species fell within the corresponding error bars.

Fig. 3.3. Measured sphericity values of glass, aluminum oxide and plastic particles

3.1.4 Preparation of Particle Size Distributions (PSDs)

The prevalent lognormal particle size distributions (PSDs) were investigated. The equation for generating the lognormal distribution is:

$$f_m = \frac{1}{d_p \sigma \sqrt{2\pi}} e^{\left[-\frac{(\ln d_p - d_{sm})^2}{2\sigma^2} \right]} \quad (3.2)$$

where f_m is the mass-based frequency distribution, d_p is the particle diameter, σ is the standard deviation and d_{sm} is the Sauter-mean diameter. Fig. 3.4 shows the exact curves obtained from Eq. (3.2), along with the discrete experimental data points, for each particle size distribution (PSD) width investigated. Experimentally, the mass percentage of each representative d_p is obtained by multiplying the f_m value with the difference between the two sieve openings of the corresponding sieve cut. In this work, the d_{sm} value of each mixture was fixed at 327.5 μm , which is approximately the middle value of the Geldart Group B size range and average of two available sieves (namely, 300 μm and 355 μm). The width of a PSD is defined dimensionlessly

as the ratio of the standard deviation (σ) to d_{sm} of the PSD (Chew et al., 2010). Lognormal PSDs with σ/d_{sm} values of 3%, 15%, 30%, 45%, 55% and 65% were examined. The narrowest PSD ($\sigma/d_{sm} = 3\%$) is such that it is approximately monodisperse, while the widest ($\sigma/d_{sm} = 65\%$) is such that it is still predominantly Group B but up to 15 mass % of Group D particles has to be included.

Prior to making up the stipulated PSDs, the glass, plastic and aluminum oxide particles obtained were first sieved using Cole-Parmer U.S. standard test sieves with diameter and height of 20.3 cm and 5.1 cm, respectively. To ensure accuracy in sieving, the sieving duration implemented was at least 1.5 times longer for the non-spherical plastic and aluminum oxide particles than the spherical glass particles.

Fig. 3.4. Lognormal frequency distributions with widths of $\sigma/d_{sm} = 15\%$, 30% , 45% , 55% and 65%

3.1.5 Procedure to measure minimum pickup velocity

The ‘weight loss method’ (Kalman et al., 2005) was used to determine pickup velocity (U_{pu}). Specifically, U_{pu} is defined as the velocity at which the mass loss due to entrainment becomes non-zero. Considering the use of acrylic for the setup, and the acknowledged electrostatic charging of the glass and plastic particles used, electrostatic charging warrants mention. Notably, electrostatic effects in particle systems have been found to be negligible when the relative humidity (RH) is above 40 % (Ciborowski and Wlodarski, 1962; Guardiola et al., 1996; Tardos and Pfeffer, 1980). Since the RH in Singapore (where the experiments were conducted) averages 80 % year-round, electrostatics effects are presumably negligible. Visual observations also attest to the lack of electrostatic charging.

First, the particle compartment of Section B was filled with the particle sample, then the mass of the bottom half of Section B was measured. The mass of glass beads needed to fill the particle compartment in Section B to the brim was determined to be 4 g, which was consistently used as the total mass of each particle sample investigated. The corresponding mass for plastic was 1.8 g and that for aluminum oxide was 5 g. The mass % values corresponding to each sieve cut (Fig. 3.4) were first determined. If these values did not add up to 100%, they were normalized accordingly. Then the actual mass of each sieve cut was determined with respect to the total mass for each species i.e. 4 g for glass, 1.8 g for plastic and 5 g for aluminum oxide. The sample was prepared by carefully weighing the corresponding mass for each sieve cut determined via the procedure elucidated above and mixing thoroughly on a weighing paper. Table 3.2 shows the details involved in preparing the glass PSD with $\sigma/d_{sm} = 30\%$. Especially in view of the small sample size and the presence of a particle size distribution, a Mettler-Toledo balance with four decimal places in the gram unit was used to prepare the samples to enhance precision. The reasonable reproducibility of the U_{pu} values measured attest to the fidelity in the sample preparation. The prepared sample was then

transferred to the chamber in the bottom part of Section B directly from the weighing paper. It was leveled by means of a small piece of smooth card-paper. All these steps were carried out with extreme care to prevent any spillage.

Table 3.2. Details of the sample preparation of the lognormal PSD of glass with $\sigma/d_{sm} = 30\%$.

Sieve mesh no.	Particle diameter range (μm)	Average diameter of sieve cut (μm)	f_m (-)	Mass %	Normalized mass %	Mass (based on 4000 mg) (mg)	Cumulative mass (mg)
140	106-150	128	0.0000375	0.16515	0.17	6.8	6.8
100	150-212	181	0.000698	4.326336	4.38	175.2	182
70	212-250	231	0.002369	9.000482	9.11	364.4	546.4
60	250-300	275	0.003716	18.58115	18.81	752.4	1298.8
50	300-355	327.5	0.004097	22.53289	22.81	912.4	2211.2
45	355-425	390	0.00317	22.18862	22.46	898.4	3109.6
40	425-500	462.5	0.001754	13.1534	13.32	532.8	3642.4
35	500-600	550	0.00068	6.802216	6.89	275.6	3918
30	600-710	655	0.000184	2.023418	2.05	82	4000
			Total	98.77366	100	4000	

Subsequently, the various sections were connected as shown in Fig. 3.1 via the screws on the flanges to ensure air-tightness. Then, the air flow to the experimental setup was set at the desired value for 120 s, which was ascertained as the duration beyond which negligible weight loss occurs. As pointed out by Tay et al. (2012), U_{pu} is theoretically independent of air flow duration. Furthermore, similar to the preliminary experimental setup validation carried out by Goy et al. (2011), the impact of air flow duration was found to be minimal at small mass losses in this work; hence the same air flow duration of 120 s (which falls in the range previously reported ((Goy et al., 2011))) was consistently used in all investigations. After that,

the air flow was shut off and the middle section weighed again to ascertain the mass loss of the particle sample. This procedure was repeated at discrete increasing values of gas velocity (U) and the mass loss at each U was recorded.

Fig. 3.5 shows an example of a mass loss versus air velocity (U) graph, whereby each datapoint represents the individual (not cumulative) mass loss at the corresponding U value. There are no error bars, since each datapoint represents exactly the mass loss occurring at the corresponding U ; the three runs in Fig. 3.5 represent the uncertainty in mass loss measurement. Specifically, each datapoint in a run was obtained by successively increasing U in steps and noting the mass loss at each step by the method described in the previous section. Three such runs were conducted in succession (not parallelly) to ensure reproducibility and also aid normalize the effects of inherent segregation. U_{pu} was determined as the U value at which the mass loss is greater than 0.01 g, and the average value of the three runs was considered. In other words, physically, each run represents an attempt to successively increase U to begin entrainment from the sample and the threshold mass loss signifies its onset. It should be noted that U_{pu} is by definition the U value corresponding to zero mass loss, thus the method used here necessitates higher U_{pu} values, but the trends with respect to the two key parameters (namely, PSD width and particle sphericity, ϕ) are expected to hold. In particular, the threshold value of 0.01 g was used instead for two reasons: (i) the extrapolation of the mass loss curve to the x -axis to determine the U value corresponding to zero mass loss is subject to error, since the assumption of linearity close to the x -axis does not strictly hold, and (ii) it is observed in Fig. 3.5 that the variability of the U value corresponding to zero mass loss is high at up to 20% of the average value of repeated tests, which is in part attributed to the inherent segregation effects. Therefore, to enable a reasonable comparison among the U_{pu} values, setting a threshold of 0.01 g not only reduces the variability in the measured values of U_{pu} but also embodies the variations in segregation. An even larger critical mass loss value than 0.01 g could be used to

better represent the segregation effects, but would undermine the objective of determining the minimum pickup velocity (U_{pu}) values. Although a logical means of determining the extent of segregation effects in the entrained sample is to measure the PSD of the remnants in the system, such a determination via sieving is not feasible due to the small size of the sample. Notwithstanding the preparation of the initial sample also involves sieving, large amounts were sieved before precisely measuring the amounts of each species required with a balance. Another means to qualify segregation could be the use of a video camera to extract information on the PSD of the entrained particles; unfortunately, the limited magnification and resolution of the current video camera restricted such quantification. Nonetheless, it should be noted that the focus in this work is on U_{pu} values characteristic of continuous PSDs and not segregation, and the PSD of the picked-up sample cannot be expected to be exactly the same as the PSD sample prepared due to the inevitable segregation effects.

Fig. 3.5. Mass loss versus superficial air velocity (U) for glass with a PSD width of $\sigma/d_{sm} = 3\%$. There are no error bars, since each datapoint represents exactly the mass loss occurring at the corresponding U .

3.2 Effect of width of particle size distribution (PSD)

Fig. 3.6 presents the pickup velocities (U_{pu}) of the three materials, namely, glass, aluminum oxide and plastic, versus the width of the lognormal PSD (σ/d_{sm}). The U_{pu} values of the monodisperse systems ($\sigma/d_{sm}=3\%$) of each material differ, because although the Sauter-mean particle diameter (d_{sm}) of each material is fixed at the same value of $327.5\ \mu\text{m}$, the particle densities (ρ_p) vary (Table 3.1). The particle sphericity (ϕ) values of the three species also vary (Table 3.1). Recall that when d_p and ρ_p are same, non-spherical particles have a greater U_{pu} than spherical ones due to the particle-particle interlocking present in the former, which takes a higher gas velocity to disengage and entrain them (Hayden et al., 2003). Here, however, the inertial effects due to varying ρ_p take precedence over the shape effects due to varying ϕ , hence the latter are not directly evident by looking at the U_{pu} values in Fig. 3.6.

Fig. 3.6 Minimum pickup velocity of various PSD widths of the three materials investigated

Surprisingly, for all three materials, U_{pu} displays a non-monotonic behavior (namely, decreases then increases) as σ/d_{sm} increases, which is counter to expectations of a binary system of Geldart Group B particles, whereby increased disparity between the non-cohesive particle species increases U_{pu} (Goy et al., 2011). Specifically, the U_{pu} values of narrower PSDs ($\sigma/d_{sm} \leq 30\%$) are lesser whereas those of wider PSDs ($\sigma/d_{sm} > 30\%$) are greater than those for monodisperse ($\sigma/d_{sm}=3\%$), with the minimum U_{pu} value occurring at $\sigma/d_{sm}=15\%$. It should be noted that such non-monotonic behavior with respect to σ/d_{sm} has been experimentally observed in segregation trends in bubbling fluidized beds (Chew et al., 2010). In particular, the extent of segregation in the bubbling fluidized bed of Geldart Group B sand particles increases then decreases as the PSD width increases, with the maximum extent of segregation observed at $\sigma/d_{sm}=30\%$. Although different physical principles likely govern the U_{pu} in horizontal pneumatic conveying and segregation extents in bubbling fluidized beds, the non-monotonic behavior in gas–solid flow processes caused by varying PSD widths warrants more understanding.

Regarding the non-monotonic U_{pu} trends with respect to σ/d_{sm} , three plausible explanations can be hypothesized, all of which are tied to the representative particle size entrained for various widths of PSDs (σ/d_{sm}). The first hypothesis is associated with the packing of the various particle size species. For narrower PSDs ($\sigma/d_{sm} \leq 30\%$), the particles smaller than d_{sm} are not small enough to occupy the interstitial voids of the particles larger than d_{sm} (Goy et al., 2011); consequently, the particles smaller than d_{sm} are more easily entrained, thereby leading to a lower U_{pu} than the monodisperse ($\sigma/d_{sm}=3\%$) sample wherein the particles are uniformly of diameter d_{sm} . As for wider PSDs, the presence of increasingly smaller and larger particles than d_{sm} implies more of the former enter the interstices of the latter, thereby the representative particle size entrained increases; thus, larger U_{pu} values are obtained. This can be verified on the basis of tap density experiments performed by Goy et al. (2011) on

binary mixtures, which revealed that the packing efficiency of the mixtures was lower than that for the monodisperse species until $d_{p,large}/d_{p,small}$ (where $d_{p,large}$ and $d_{p,small}$ were the particle diameters of the larger and small species, respectively) was ≈ 4 ; beyond which it started rising. A low packing efficiency implied that the smaller particles were not able to enter the interstitial voids of the larger ones, resulting in individual particle pickup lowering the U_{pu} . In case of the lognormal PSDs tested here, it can be seen from Fig. 3.4 that the PSD with $\sigma/d_{sm}=15\%$ does not have any particles with $d_{p,large}/d_{p,small} > 4$. For PSDs with $\sigma/d_{sm} \geq 30\%$, the proportion of such particles increases with increasing PSD width, thereby allowing more amounts of smaller particles to enter the interstitial voids of the larger ones and consequently increasing U_{pu} . The second hypothesis relates to inter-particle momentum transfer among the various size species. Specifically, the more easily ‘picked up’ smaller particle may transfer their momentum to a larger one upon collision, a multiplied effect of which may cause the larger particle to be ‘picked up’. Analogously, for non-cohesive particles, the presence of smaller particles (whose terminal velocity is lesser than the operating superficial gas velocity) has been observed to enhance the elutriation of larger particles (whose terminal velocity is greater than the operating superficial gas velocity) in gas–solid fluidization (Chew et al., 2013; Geldart et al., 1979). On one hand, for the narrower PSDs ($\sigma/d_{sm} \leq 30\%$), part of the higher momentum of particles smaller than d_{sm} is transferred to the larger particles; hence entrainment of the particle sample is facilitated thereby lowering the U_{pu} . On the other hand, for the wider PSDs ($\sigma/d_{sm} > 30\%$), the increasing range of particle sizes implies increasing discrepancy between the minimum and maximum particle sizes, which impede adequate momentum transfer among the various particle species to enable entrainment. The third hypothesis relates to varying extents of species segregation. As has been noted in the previous section, inherent segregation may already be present in the samples even before entrainment occurs, as seen from the variation (up to 20%) at zero mass loss (Fig. 3.5). Though taking an average of multiple runs and recording U_{pu} at

0.01 g weight loss, where variability was less will help normalize segregation effects for a PSD width, varying degrees of segregation for different PSD widths, which has been noted before (Chew et al., 2011a; Chew et al., 2013; Chew et al., 2010) could account for the non-monotonic effects especially in view of the non-monotonic extents of segregation previously reported in a bubbling fluidized bed (Chew et al., 2010).

The impact of PSD width is further shown in Fig. 3.7, which shows a plot of Re_p^* [Eq. (2.5)] versus Ar [Eq. (2.6)] obtained by empirical correlations developed for monodisperse particles (Kalman et al., 2005), and binary and ternary particle mixtures (Tay et al., 2012). Note that a correlation for continuous PSDs is not available. Also displayed are the data generated for the three materials at various PSD widths in this work. While the Ar values for each material are the same (because the d_{sm} and ρ_p values are constant for a given material), the Re_p^* values vary with the U_{pu} values of the various PSD widths.

Fig. 3.7. Comparison of the relationship between Re_p^* vs Ar given by monodisperse (Kalman et al., 2005), and binary and ternary (Tay et al., 2012) correlations and for experimental results from this work.

The spread of data points due to PSD width variations indicates the non-negligible impact of PSD on U_{pu} . The scatter is greatest for plastic (least spherical) and least for glass beads (most spherical), presumably because the empirical correlations are inadequate to incorporate the combined effects of both PSD width and ϕ . For the materials investigated here, better agreement with both the Kalman et al. (2005) and Tay et al. (2012) correlations is observed for both the monodisperse ($\sigma/d_{sm}=3\%$) and the intermediate PSDs ($\sigma/d_{sm}=45\text{--}55\%$). It should be noted that the effect of PSD width was not considered in the development of both correlations and only the size ranges of the particles (but not the actual PSD) were reported. The spread of experimental data due to PSD widths highlights the need to report and incorporate effects of the nature of the PSDs investigated. Notably, whereas the empirical correlations predict that U_{pu} is higher for polydisperse than monodisperse systems at lower Ar values (i.e., for plastic) and vice versa at higher Ar values (i.e., for glass and aluminum oxide), experimental results in this work show that the non-monotonic U_{pu} trend with respect to σ/d_{sm} is consistent across the range of Ar values considered (i.e., for all three materials). The discrepancy between empirical correlations and experimental data could be due to the lack in the former of (i) mechanistic understanding in such empirical correlations, which restrict the application beyond the specific system tested (Breault, 2006; Chew et al., 2015), and/or (ii) consideration of detailed variations of PSD widths and shape effects per studied in this work, which highlights the need for more understanding in these respects.

3.3 Effect of particle shape

To examine the impact of particle shape, the extent of non-monotonic behavior of U_{pu} with respect to σ/d_{sm} among the three materials is compared. Notably, differences in particle sphericity (ϕ) lead to variations in (i) particle rotation and (ii) particle lift (Hölzer and Sommerfeld, 2009; Zastawny et al., 2012), both of which impact U_{pu} values. Correspondingly,

the extent of non-monotonic behavior gives an indication of the impact of particle shape, because the higher representation of the larger particle sizes in the wider PSDs enhances the particle rotation and lift effects (Lee and Hsu, 1996). The U_{pu} values for each material are normalized with respect to the U_{pu} of the monodisperse system ($\sigma/d_{sm}=3\%$) to facilitate a more direct contrast, as shown in the normalized plot of U_{pu} values in Fig. 3.8. The y-axis represents the normalized U_{pu} (i.e., $U_{pu}/U_{pu,mono}$), while the x-axis represents σ/d_{sm} . As apparent in Fig. 3.8, the increase in U_{pu} from $\sigma/d_{sm}=15\%$ to $\sigma/d_{sm}=65\%$ width is greatest for the most non-spherical particles (namely, plastic with $\varphi=0.75$) and least for the most spherical particles (namely, glass with $\varphi=0.97$). In other words, the lower the particle sphericity (φ) is, the more extensive the non-monotonic behavior becomes, which calls for a closer inspection of the impact of particle rotation and lift in lowering U_{pu} at intermediate PSD width ($\sigma/d_{sm}=15\text{--}30\%$) and amplifying U_{pu} at wider PSD ($\sigma/d_{sm}>30\%$) relative to the monodisperse ($\sigma/d_{sm}=3\%$) system. It should be noted that although particle rotation and particle lift effects typically relate to suspended particles, the incipient entrainment that characterizes U_{pu} is possibly subject to such effects too (Cabrejos and Klinzing, 1994; Zenz, 1964), which hence provide plausible reasons underlying the different extents of non-monotonic behaviors among particles of different sphericities (φ).

Fig. 3.8. Normalized minimum pickup velocity of various PSDs of three materials

Regarding particle rotation, Fig. 3.9 shows frames of plastic particles ($\phi=0.75$) taken using a high-speed camera at 1 ms intervals, which clearly shows the rotation of the non-spherical particle as it travels. The term ‘coefficient of moment’ warrants mention. Specifically, the coefficient of moment of a particle is defined as the ratio of the torque experienced by the particle to the kinetic energy of the fluid displaced by that particle (Hölzer and Sommerfeld, 2009). On one hand, for a spherical particle, the coefficient of moment of the particle at rest in a moving fluid is zero (Hölzer and Sommerfeld, 2009; Zastawny et al., 2012); hence it maintains a stable position. On the other hand, for a non-spherical particle, the coefficient of moment is non-zero for all except one orientation; hence it is in an unstable position because the torque about its axis causes it to rotate. Note that spherical particles may also undergo rotation about their axes, but as these particles are completely symmetrical in three dimensions,

their coefficient of moment is zero and there is no effect of the torque on their motion as opposed to their non-spherical counterparts. Consequently, the effects of particle rotation are more visible and enhanced in non-spherical particles ($\phi < 1$) than spherical ones ($\phi = 1$), due to which the corresponding drag force is inevitably increased in the former compared to the latter. This effect is further accentuated by increasing Reynolds number (Njobuenwu and Fairweather, 2013) and has been reported to be significant at Reynolds number greater than 30 (Hölzer and Sommerfeld, 2009); note the experiments in this work were carried out at Reynolds number greater than 80. For wider PSDs ($\sigma/d_{sm} > 30\%$) of non-spherical ($\phi < 1$) particles, because the air supplied has to overcome the increased drag due to more extensive rotation associated with the larger particles (Lee and Hsu, 1996), the U_{pu} value is correspondingly more amplified.

Fig. 3.9. High-speed camera images of plastic showing particle rotation at 1 ms intervals. The air flow is from left to right in all frames.

With respect to particle lift, recall that the lift force acts perpendicular to the fluid flow. The lift coefficient, defined as the ratio of the lift force to the kinetic energy of the fluid displaced

by the particle, is zero for spherical particles ($\varphi=1$) and non-zero for non-spherical particles ($\varphi<1$) (Hölzer and Sommerfeld, 2009; Zastawny et al., 2012). Similar to the effect of particle rotation, some of the air supplied has to be expended in overcoming the particle lift of the non-spherical particle in the direction perpendicular to fluid flow. As a result, due to the enhanced effect of particle lift associated with the larger particles (Lee and Hsu, 1996) in wider PSDs ($\sigma/d_{sm} > 30\%$), the U_{pu} value of non-spherical particles ($\varphi<1$) increases relative to that of spherical ones ($\varphi=1$).

Furthermore on particle sphericity, a modification to Fig. 3.7 in terms of re-defining the x -axis with a modified Archimedes number (Ar^* has been presented by Kalman et al. (2005))

$$Ar^* = 0.03Ar e^{3.5\varphi} \quad (3.3)$$

whereby Ar^* is the Archimedes number modified for particle sphericity calculated using Eq. (3.3) and Ar is the measured Archimedes number determined using Eq. (2.6). Specifically, Eq. (3.3) was derived by curve-fitting of experimental data on a plot of Ar^*/Ar versus φ and implies that lower φ values depresses the characteristic Ar (i.e., Ar^*). Fig. 3.10 shows a plot of Re_p^* vs Ar^* , whereby Re_p^* is the particle Reynolds number modified to account for the pipe diameter as a ratio of a reference diameter (Eq. (2.5)) and Ar^* is the Archimedes number modified to account for φ (Eq. (3.3)), for all materials and PSD widths investigated. While the experimental data in this work scatter around the two correlations for the unmodified Ar (Fig. 3.7), more discrepancy is observed for the plot for Ar^* (i.e., Ar corrected for sphericity based on Eq. (3.3)) in Fig. 3.10. Ironically, the correlations appear to perform worse for the non-spherical particles (i.e., aluminum oxide and plastic), presumably because (i) the non-spherical particles investigated by Kalman et al. (2005). belongs to Geldart Group D, and/or (ii) the empirical correlation is inadequate to incorporate the combined effects of both PSD and φ . The worst agreement between the experimental data of all there materials and both the Kalman et

al. (2005) and Tay et al. (2012) correlations is observed for the widest PSDs ($\sigma/d_{sm} = 55 - 65\%$), which again highlights the need to incorporate the effects of PSD widths in the development of such correlations.

Fig. 3.10. Re_p^* versus Ar^* (Kalman et al., 2005), where Re_p^* is the particle Reynolds number modified to account for the pipe diameter as a ratio of a reference diameter (Eq. (2.5)) and Ar^* is the Archimedes number modified to account for particle sphericity (Eq. (3.3)).

3.4 Conclusions

Experiments were conducted to study the impact of particle size distribution (PSD) and particle sphericity (ϕ) on the minimum pickup velocity (U_{pu}). Three materials of different ϕ and ρ_p were investigated at PSD widths (σ/d_{sm}) spanning 3% (approximately monodisperse) to 65%. On the impact of PSD, consistently for all three materials, U_{pu} consistently trends non-monotonically (namely, decreases then increases) with σ/d_{sm} , and the minimum U_{pu} occurs for PSDs with $\sigma/d_{sm}=15\%$. Regarding particle sphericity, the extent of

non-monotonic behavior is greater for particles with lower ϕ , due to the more dominant effects of particle rotation and particle lift.

The implications of PSD width and ϕ revealed here are applicable to gas–solid systems in general, and are expected to find additional value in the validation of models. The experimental data and understanding presented here can aid in enhancing the ability of two-fluid models to adapt the various closures (kinetic theory, drag laws, etc.) to continuous PSDs and particle sphericity.

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4

Effect of particle diameter, density and shape on minimum pickup velocity of binary mixtures

This chapter discusses the impacts of particle diameter (d_p), particle density (ρ_p) and particle sphericity (ϕ) on the minimum pickup velocity (U_{pu}) of binary mixtures of Geldart group B particles.

4.1 Experimental protocol

The experimental unit and the method to determine U_{pu} used here are identical to those used in the previous study. The reader is referred to Sections 3.1.1 and 3.1.5 for details on the experimental unit and the procedure to measure U_{pu} respectively.

4.1.1 Materials

The solids particles used were glass, plastic and aluminum oxide from Abrasive Engineering Pte. Ltd., Singapore. Each particle type was carefully sieved to obtain the required particle diameter (d_p) required. Solids of three d_p values of each particle type was investigated. The particle sphericity (ϕ) was determined by the projected image technique (Section 3.1.3). The solids particles in this study belonged to Geldart group B (Geldart, 1973), and they were so chosen specifically to eliminate inter-particle cohesive effects. The particle properties particle diameter (d_p), particle density (ρ_p), and particle sphericity (ϕ) of the solids tested are given in Table 4.1. For easy reference glass, aluminum oxide and plastic are termed ‘G’, ‘A’ and ‘P’, respectively, and the d_p values of 181 μm , 327.5 μm and 550 μm are referred to as ‘181’, ‘327’ and ‘550’, respectively. For example, glass particles with a d_p value of 181 μm , is referred to as ‘G181’.

Table 4.1. Properties of the particle species used in this study.

Species	Material	Particle diameter, d_p (μm)	Label	Particle density, ρ_p (kg/m^3)	Sphericity, ϕ (-) (Anantharaman et al., 2015)	Minimum pickup velocity, U_{pu} (m/s)
G181	Glass	181 $\pm$ 31	G181	2500	0.97	4.78
G327		327.5 $\pm$ 27.5	G327			5.49
G550		550 $\pm$ 50	G550			6.55
A181	Aluminum Oxide	181 $\pm$ 31	A181	3920	0.82	5.13
A327		327.5 $\pm$ 27.5	A327			5.84
A550		550 $\pm$ 50	A550			7.26
P181	Plastic	181 $\pm$ 31	P181	1150	0.74	3.01
P327		327.5 $\pm$ 27.5	P327			3.72
P550		550 $\pm$ 50	P550			4.78

The three binary mixtures investigated were of three different types; binary-size (i.e., the two constituents differed only in d_p), binary-density (i.e., the two constituents differed only in ρ_p), and binary-size-density (i.e., the two constituents differed in both d_p and ρ_p). For each binary mixture, the pickup velocities for the two monodisperse constituents, together with those of mixtures of three percent volume fractions 25, 50 and 75 percent volume of each constituent, were measured. Since the mass required to fill up the entire volume of the chamber in the bottom part of Section B of the apparatus (Fig. 3.1) was known (4 g for glass, 5 g for aluminum oxide and 1.8 g for plastic) from Section 3.1.5, volume fractions of the constituents of the binary mixtures were equal to the corresponding mass fractions. Hence, for example, to prepare a 25% mixture of P181-A327, 1.25 g of the 327.5 μm aluminum oxide was mixed with 1.35 g

of the 181 μm plastic. The steps after preparation of the mixture sample were the same as described in Section 3.1.5.

4.2 Minimum pickup velocity of binary-size, binary-density and binary-size-density mixtures

4.2.1 Binary-size mixtures

Fig. 4.1a shows the pickup velocities (U_{pu}) of four binary-size mixtures as a function of volume percent of the larger ($d_p = 550 \mu\text{m}$) species. Fig. 4.1b shows the analogous plot for U_{pu} normalized with the U_{pu} value of the smaller species ($U_{pu}/U_{pu,small}$) to allow for a more straightforward comparison of the various binary-size mixtures despite the different U_{pu} values. The four mixtures are (i) glass with 181 μm particles and 550 μm particles (G181–G550), (ii) aluminum oxide with 181 μm particles and 550 μm particles (A181–A550), (iii) plastic with μm particles and 550 μm particles (P181–P550), and (iv) plastic with 181 μm particles and 327 μm particles (P181–P327).

Fig. 4.1 (a) U_{pu} and (b) U_{pu} normalized by the U_{pu} of the smaller species ($U_{pu}/U_{pu,small}$) versus volume percent of the larger species of four binary-size mixtures. The dashed lines in (a) join the U_{pu} values of the two constituents in the respective binary-size mixtures.

Fig. 4.1a shows as expected, that the monodisperse aluminum oxide particles had the highest U_{pu} values due to their highest particle densities (ρ_p). Likewise the plastic particles had the lowest U_{pu} values due to their least particle densities (ρ_p) (Cabrejos and Klinzing, 1994). Also, U_{pu} increased as the volume percent of the larger species increased, but only up to 75 vol%, beyond which the mixture U_{pu} became equal to that of the larger constituent, which indicates the dominance of the larger constituent in dictating mixture U_{pu} values. Interestingly, although the previous work (Goy et al., 2011) reported that U_{pu} increased linearly with increasing volume fraction for binary-size mixtures if both species belong to Geldart Group B, all the trends in Fig. 4.1a were non-linear, with the P181–P550 plastic particles displaying the most deviation from linearity. The non-linear variation here is presumably because these plastic particles were the least spherical (Table 4.1), whereas perfect spheres were used in the previous study (Goy et al., 2011). The other binary-size plastic mixture consisting of a smaller d_p differential, P181–P327 had a lesser deviation from linearity than the P181–P550 mixture due to the lesser particle lift and rotation effects associated with smaller particles (refer Section 3.3).

In Fig. 4.1b, the non-linearity effect due to particle sphericity is more clear. The most non-linear trend was for P181–P550, followed by A181–A550, then G181–G550, and finally P181–P327. Previous studies have indicated that non-spherical Geldart group B particles exhibited a greater degree of non-monotonicity in U_{pu} with respect to PSD width (refer Section 3.3), which is due to the greater particle rotation and lift associated with less spherical particles. In that case the pickup velocity of the mixture tended to mimic that of the larger species more, which caused its value to lie above the linear line drawn between the U_{pu} values of the two monodisperse constituents. Here ‘mimic’ refers to the fact that the U_{pu} values of the mixture at the different volume percent tended to be much closer to that of one constituent than that of the other. The more dominant role of the larger species on the U_{pu} value was explained (Lee and

Hsu, 1996) to be at least in part due to the increased particle rotation and lift effects associated with larger, non-spherical particles. The twin effects of least sphericity of the plastic particles (P) and larger d_p of the larger constituent (in the case of P181–P550 relative to P181–P327) hence provide a cogent explanation for the most non-linear variation of U_{pu} with volume percent of the larger species exhibited by the P181–P550 plastic binary-size mixture in Fig. 4.1a and b.

4.2.2 Binary-density mixtures

Fig. 4.2 shows the measured pickup velocities (U_{pu}) of the six binary-density mixtures. Fig. 4.2a shows the U_{pu} values as a function of the volume percent of the denser species and Fig. 4.2b shows U_{pu} normalized with respect to the U_{pu} of the less dense species ($U_{pu}/U_{pu,less\ dense}$) also as a function of the volume percent of the denser species. It should be noted that (i) the U_{pu} values of G181 and P550 were the same at 4.78 m/s (Table 4.1), hence the datapoints representing these two species coincide on the y-axis of Fig. 4.2a, and (ii) the difference in the U_{pu} values of G181 and A181 (Table 4.1) was the same as the error involved in the measurement, hence the quantification of the mixture U_{pu} values was not carried out and a linear line joining the U_{pu} values of its constituents was instead used in Fig. 4.2 as a reasonable representation.

Fig. 4.2. (a) U_{pu} and (b) U_{pu} normalized by the U_{pu} of the less dense species ($U_{pu}/U_{pu,less\ dense}$) versus volume percent of the denser species of six binary-density mixtures. The dashed lines in (a) join the U_{pu} values of the two constituents in the respective binary-density mixtures.

Three observations in Fig. 4.2a are as expected. First, because aluminum oxide particles were the densest species investigated, the largest A550 monodisperse particle type gave the highest U_{pu} value. Second, because aluminum oxide particles were denser than glass particles, the U_{pu} values of the binary-density mixtures of plastic with aluminum oxide (P181-A181 or P550-A550) were greater than those of the binary-density mixtures of plastic with glass (P181-G181 or P550-G550). Thirdly, the U_{pu} values for the binary-density mixture containing the larger constituents (e.g., P550-A550) were greater than those containing the smaller constituents (e.g., P181-A181). Another observation regarding the comparison between the binary-size (Fig. 4.1) and binary-density (Fig. 4.2) mixtures is worth noting. Similar to the binary-size mixtures in Fig. 4.1, the U_{pu} values of the 50 and 75 vol% mixtures were all above the linear trendline connecting the U_{pu} values of the two constituent species, which suggest a more dominant role of the denser constituent in the binary-density mixture. In contrast to the more linear U_{pu} variation for binary-size in Fig. 4.1, the trends for the binary-density mixtures were such that the U_{pu} value at 25 vol% in all cases was approximately on the linear trendline, which implied that the shape effects resulting in non-linear trends were negated at 25 vol% for binary-density mixtures.

In the normalized U_{pu} plots in Fig. 4.2b, two observations are worth highlighting. Firstly, the trends of P181-G181 and P181-A181 were similar, while that of P550-G550 and P550-A550 were similar. This indicates that the different sphericity (ϕ) values of the denser constituent of glass (G) and aluminum oxide (A) in their binary-density mixtures with plastic (P) did not influence the profiles significantly. Secondly, with regards to the effect of d_p in binary-density mixtures, contrasting trends were observed. On one hand, the $U_{pu}/U_{pu,less\ dense}$ values of the smaller P181-G181 and P181-A181 were greater than the larger P550-G550 and P550-A550, respectively. On the other hand, the $U_{pu}/U_{pu,less\ dense}$ values of the smaller

G181-A550 was in contrast lower than that of the larger G550-A550. The contrast between P-G and G-A was especially surprising, since the difference in ρ_p values of the constituent species was similar. A plausible underlying reason may be related to the particle sphericity (ϕ) of the more entrainable species (i.e., the constituent species with lower ρ_p). The species with the lower U_{pu} was P (i.e., the least spherical species) for the case of P-G and P-A, but G (i.e., the most spherical species) for the case of G-A. For P-G and P-A, due to the increased particle rotation and lift associated with the larger (i.e., $d_p = 550 \mu\text{m}$) P species (Refer Section 3.3), the $U_{pu}/U_{pu,less\ dense}$ ratio was lower for the larger species. On the contrary for G-A, in the absence of particle rotation and lift for G, the $U_{pu}/U_{pu,less\ dense}$ ratio was higher for the larger species (i.e., $d_p = 550 \mu\text{m}$) due to the greater associated inertia. This is in sync with the earlier section on binary-size mixtures (Fig. 4.1), whereby plastic (P) showed a greater relative increase in U_{pu} with an increase in d_p than aluminum oxide (A) due to the enhanced particle rotation and lift effects in the former.

4.2.3 Binary-size-density mixtures

In binary-size-density mixtures, the two constituent species differ in both particle size (d_p) and particle density (ρ_p). Two cases can clearly be defined here: (i) the larger-sized constituent is the denser one, and hence has a higher pickup velocity (U_{pu}), and (ii) the larger-sized constituent is the less dense one and has a lower pickup velocity (U_{pu}).

Fig. 4.3a shows the pickup velocities (U_{pu}) of three binary-size-density mixtures belonging to case (i), wherein the larger constituent is the denser one, as a function of the volume percent of the species with the larger pickup velocity (U_{pu}). Fig. 4.3b represents the same U_{pu} values normalized by the pickup velocity (U_{pu}) of the constituent with the lower U_{pu} ($U_{pu}/U_{pu,lower}$). Two observations are noted. Firstly, comparing the trends in Fig. 4.3a for G181-A327 and P181-A327, although glass and plastic particles differed significantly in

particle densities, the U_{pu} values of the two binary mixtures were very similar for volume percent $\geq 50\%$. This indicates the dominant role of the aluminum oxide particles with $d_p = 327 \mu\text{m}$ (A327) in influencing U_{pu} in this composition range, which is consistent with the observations made above of the larger or denser particle constituent being more dominant. Secondly, the non-linear variations were more similar to that of a binary-size mixture than a binary-density mixture. Recall that the U_{pu} values for 25 vol% binary-density mixtures lay on the linear trendline connecting the U_{pu} values of the two constituent species (Fig. 4.2a). This implies that, between d_p and ρ_p , d_p was the more dominant parameter.

Fig. 4.3 (a) U_{pu} and (b) U_{pu} normalized by the U_{pu} of the species with the lower U_{pu} ($U_{pu}/U_{pu,lower}$) versus volume percent of the species with higher U_{pu} of three binary-size-density mixtures. In all three binary-size-density mixtures, the larger-sized constituent had a higher ρ_p . The dashed lines in (a) join the U_{pu} values of the constituent species in each binary mixture.

Fig. 4.4a shows the pickup velocities (U_{pu}) of three binary-size-density mixtures as a function of the volume percent of the species with larger U_{pu} . The constituent with the higher U_{pu} was smaller and denser, corresponding to case (ii). Fig. 4.4b shows U_{pu} values normalized by the U_{pu} of the constituent with lower U_{pu} . Two surprising observations inconsistent with our observations so far are worth highlighting. Firstly, whereas all the above U_{pu} trends have been non-linear, the P327-A181 mixture exhibited a linear variation,

which was observed in a previous study (Goy et al., 2011) for binary-size mixtures of spherical particles. It appears that for the unique combination of P327 and A181 particles, the impact on minimum pickup velocity (U_{pu}) due to particle sphericity was negated, or more specifically, the inertial forces were balanced by rotation and lift effects caused by the non-spherical plastic and aluminum oxide particles. Secondly, whereas all the above U_{pu} trends were such that the constituent with the greater U_{pu} had a more significant influence on the U_{pu} values of the binary mixture, the constituent with the lower U_{pu} was more dominant in the P327-G181 and P550-A327 binary mixtures, as all their U_{pu} values are lying below the linear lines connecting the U_{pu} values of the individual constituents. Despite this observation being seemingly contradictory, the common thread here is that d_p influenced U_{pu} more than ρ_p . The lower U_{pu} values of the larger-sized non-spherical plastic particles influenced the mixture U_{pu} values more than the higher U_{pu} values of the smaller-sized glass and aluminum oxide particles.

Fig. 4.4 (a) U_{pu} and (b) U_{pu} normalized by the U_{pu} of the species with lower U_{pu} versus volume percent of the species with the higher U_{pu} of three binary-size-density mixtures. In all three mixtures, the smaller-sized constituent had a higher ρ_p , and was the constituent with the higher U_{pu} . The dashed lines in (a) join the U_{pu} values of the constituent species in each binary mixture.

The more dominant influence of the larger constituent on U_{pu} may be linked to the Brazil nut phenomenon (Rosato et al., 1987), whereby the larger particles preferentially segregate to the top, hence are the ones entrained at U_{pu} . However, the same is not strictly true for the denser constituent (Mobius et al., 2001). Further studies are needed for a more mechanistic understanding on the pickup of binary mixtures.

4.3 Force balance analysis to determine minimum pickup velocity

4.3.1 Representation of a non-spherical particle

Gomes and Mesquita (2013) modeled a non-spherical particle as a sphere with a segment cut at an appropriate angle (θ) with the flat face resting on the pipe wall, as shown in Fig. 4.5.

Fig. 4.5. Representation of a non-spherical particle (Gomes and Mesquita, 2013)

The projected area (A_p) of this particle is given by:

$$A_p = \frac{R^2}{2} (2\pi - \theta + \sin\theta) \quad (4.1)$$

Substituting $f_1(\theta) = (2\pi - \theta + \sin\theta)$ for brevity, A_p becomes:

$$A_p = \frac{R^2}{2} f_1(\theta) \quad (4.2)$$

Sphericity (φ) of the particle can be defined from Section 3.1.3 as:

$$\varphi = \frac{2\sqrt{A_p/\pi}}{D_{cmin}} \quad (4.3)$$

where D_{cmin} is the diameter of the minimum circumscribed circle around the projected 2-D image of the particle. Substituting $2R$ for D_{cmin} , and substituting Eq. (4.2) into Eq. (4.3) gives:

$$\varphi = \sqrt{1 - \frac{\theta}{2\pi} + \frac{\sin\theta}{2\pi}} \quad (4.4)$$

Fig. 4.6 shows a plot of θ against φ based on Eq. (4.4), which shows that each particle sphericity, φ , has a unique value of θ .

Fig. 4.6. Relationship between θ and φ , determined based on the method of Gomes and Mesquita (2013)

To obtain the relationship between the diameter of the particle investigated (d_p) and R , the volume of the particle investigated (which is considered equal to that of a sphere with diameter d_p) is equated to the volume of the model particle, which is given by subtracting the volume of the spherical cap from the volume of the sphere (Fig. 4.5)

$$\frac{\pi d_p^3}{6} = \frac{\pi R^3}{3} \left[2 + 3 \cos\left(\frac{\theta}{2}\right) - \cos^3\left(\frac{\theta}{2}\right) \right] \quad (4.5)$$

Solving for R and substituting in Eq. (4.2) gives

$$A_p = \frac{d_p^2 f_1(\theta)}{2 \left[4 + 6 \cos\left(\frac{\theta}{2}\right) - 2 \cos^3\left(\frac{\theta}{2}\right) \right]^{\frac{2}{3}}} \quad (4.6)$$

Substituting $f_2(\theta) = [4 + 6 \cos(\theta/2) + 2 \cos^3(\theta/2)]^{2/3}$ for brevity, Eq. (4.6) becomes

$$A_p = \frac{d_p^2 f_1(\theta)}{2 f_2(\theta)} \quad (4.7)$$

4.3.2 Force balance

The drag force acting on a particle initially at rest in a horizontal pneumatic conveying system with superficial velocity U is given by (Rabinovich and Kalman, 2009b):

$$F_D = n C_D A_p \frac{\rho U^2}{2} \quad (4.8)$$

where C_D is the drag coefficient (typically, 0.44) (Rabinovich and Kalman, 2009b), n is the coefficient to account for flow over a layer of particles and ρ is the density of air (1.2 kg/m³ at atmospheric conditions). The presence of a wall has an impact on the C_D of a particle. C_{DW} is the drag coefficient of a particle in a pipe of diameter D with non-negligible wall effects. Hence, to account for wall effects, Eq. (4.8) is multiplied by the ratio $C_{DW}/C_{D\infty}$ (where $C_{D\infty}$ is the drag coefficient with wall effects absent). The expression for $C_{DW}/C_{D\infty}$ given by Munroe (1888) as recommended by Cabrejos and Klinzing (1992) and Klinzing et al. (2010) is used here:

$$F_D = \frac{C_{Dw}}{C_{D\infty}} \cdot n C_D A_p \frac{\rho U^2}{2} = \left[1 - \left(\frac{d_p}{D} \right)^{1.5} \right]^{-2} n C_D A_p \frac{\rho U^2}{2} \quad (4.9)$$

Substituting the expression for projected area A_p from Eq. (4.7), F_D can be expressed as

$$F_D = \frac{1}{4} \left[1 - \left(\frac{d_p}{D} \right)^{1.5} \right]^{-2} n C_D \rho U^2 d_p^2 \frac{f_1(\theta)}{f_2(\theta)} \quad (4.10)$$

The coefficient n is used to account for pickup over a layer of particles. Cabrejos and Klinzing (1994) have done this by multiplying their expression by a correction factor, while Rabinovich and Kalman (2009b) have added an implicit correction. Gomes and Mesquita (2013) have fitted experimental data from glass and sand particles ($\rho_p = 2480 - 2834 \text{ kg/m}^3$, $d_p = 22 - 3860 \text{ }\mu\text{m}$ and pipe diameter, $D = 26 - 152 \text{ mm}$), and that from past studies (Cabrejos and Klinzing, 1994; Kalman et al., 2005) to obtain an expression for n via dimensional analysis, which is used in this model:

$$n = 0.57 \left(\frac{U \sqrt{d_p}}{D \sqrt{g}} \right)^{0.32} \quad (4.11)$$

where D is the pipe diameter.

The opposing force in the horizontal direction is the frictional force (F_f), which is given by (Cabrejos and Klinzing, 1992)

$$F_f = f(F_g - F_b) \quad (4.12)$$

where f is the coefficient of friction (typically, 0.85) (Rabinovich and Kalman, 2009b). Note that although the experimental materials used in this study were glass, plastic and aluminum oxide, the attempt is for the model to be applicable to Geldart Group B (Zone I) particles in general. Hence, the f value of sand, a typical Geldart Group B material was considered for this model, which is ~ 0.85 as determined by (Rabinovich and Kalman, 2009a). F_g is the gravitational force

$$F_g = \frac{\pi d_p^3}{6} g \rho_p \quad (4.13)$$

and F_b is the buoyancy force

$$F_b = \frac{\pi d_p^3}{6} g \rho \quad (4.14)$$

where ρ_p and ρ are particle and fluid densities, respectively. Recall that the volume of the particle investigated is considered to be the volume of a sphere with diameter d_p , which is used in Eqs. (4.13) and (4.14). Note that this volume is the same as that of the model particle as stated in Eq. (4.5). Substituting Eqs. (2.6), (4.13) and (4.14) into Eq. (4.12) gives F_f as

$$F_f = f(F_g - F_b) = \frac{\pi}{6} Ar \frac{\mu_f^2}{\rho} f \quad (4.15)$$

Horizontal motion occurs when the drag force (F_D) due to fluid flow just overcomes the frictional force (F_f). Hence, at incipient pickup (i.e., $U = U_{pu}$),

$$F_D = F_f \quad (4.16)$$

Substituting Eq. (4.10) and Eq. (4.15) into Eq. (4.16) and solving for U , the minimum pickup velocity $U_{pu,calculated}$ is obtained as

$$U_{pu,calculated} = \left[1 - \left(\frac{d_p}{D} \right)^{1.5} \right] \left[\sqrt{\frac{\frac{2\pi}{3} f Ar \frac{\mu_f^2}{\rho} f_2(\theta)}{nC_D \rho d_p^2 f_1(\theta)}} \right] \quad (4.17)$$

Fig. 4.7 shows the minimum pickup velocity (U_{pu}) as calculated by Eq. (4.17) plotted against sphericity (ϕ) for particles with $\rho_p = 2500 \text{ kg/m}^3$ and $d_p = 181 \text{ }\mu\text{m}$, $327.5 \text{ }\mu\text{m}$ and $550 \text{ }\mu\text{m}$. It is clear from Fig. 4.7 that U_{pu} decreases as ϕ increases, indicating that non-spherical particles require greater gas velocity to be entrained than spherical ones. The trend holds for particles with different ρ_p values. This agrees with previous reports (Cabrejos and Klinzing, 1994;

Gomes and Mesquita, 2013; Hayden et al., 2003; Kalman et al., 2005) regarding the general relationship between ϕ and U_{pu} .

Fig. 4.7 Calculated pickup velocity calculated per Eq. (4.17) versus sphericity for 181 μm , 327.5 μm and 550 μm particles of density 2500 kg/m^3

4.3.3 Comparison of results with model

Since the constituent species of the investigated binary mixtures had different particle properties, a suitable averaging technique must be applied to determine the characteristic properties of the mixtures. Goy et al. (2011) measured U_{pu} of binary-size mixtures of spherical glass beads and found that the mixture characteristic particle diameter ($d_{p,mix}$) can be well-represented by either the arithmetic-mean or Sauter-mean for Zone I (i.e., Geldart group B) particles, since both mean values were similar for the range of particle diameters investigated. Accordingly, for the similar range of particle diameters in Zone I investigated in the present

work, the arithmetic-mean was adopted to determine the characteristic particle diameter of the mixture:

$$d_{p,mix} = x_1 d_{p1} + x_2 d_{p2} \quad (4.18)$$

where x_1 and x_2 are the mass fractions of the constituent species with particle diameters of d_{p1} and d_{p2} , respectively. For the characteristic particle density of the binary mixtures ($\rho_{p,mix}$), because no prior effort has been carried out for horizontal pneumatic conveying, the arithmetic-mean used in fluidization studies of binary-density mixtures was adopted (Asif, 2013; Chiba et al., 1979; Di Maio et al., 2012):

$$\rho_{p,mix} = x_1 \rho_{p1} + x_2 \rho_{p2} \quad (4.19)$$

where x_1 and x_2 are the mass fractions of the constituent species with particle densities of ρ_{p1} and ρ_{p2} , respectively. No studies are available on the characteristic particle sphericity of binary-sphericity mixtures (ϕ_{mix}). As a first approximation, an arithmetic mean ϕ_{mix} was assumed:

$$\phi_{mix} = x_1 \phi_1 + x_2 \phi_2 \quad (4.20)$$

where x_1 and x_2 are the mass fractions of the constituent species with particle sphericities of ϕ_1 and ϕ_2 , respectively. The validity of Eq. (4.20) was checked with the Re_p^* versus Ar plot for Zone I particles (Kalman et al., 2005). Fig. 4.8 shows the Re_p^* vs. Ar plot of all the binary mixtures investigated here with the mixture characteristic properties $d_{p,mix}$, $\rho_{p,mix}$, ϕ_{mix} determined by Eqs. (4.18), (4.19), (4.20) respectively. The experimental data were within $\pm 15\%$ of the Zone I correlation predictions (Kalman et al., 2005), which affirms that using an arithmetic mean ϕ_{mix} is reasonable.

Fig. 4.8. Re_p^* versus Ar plot for all the binary mixtures studied here. The mixture particle diameter ($d_{p,mix}$), density ($\rho_{p,mix}$) and sphericity (ϕ_{mix}) were calculated using Eqs. (4.18) – (4.20).

With the characteristic properties of the different types of binary mixtures studied here calculated from Eqs. (4.18) – (4.20), the validity of the U_{pu} derived from force balance (Eq. (4.17)) was investigated. Fig. 4.9 shows a parity plot of $U_{pu,calculated}$ (Eq. (4.17)) versus $U_{pu,measured}$, obtained experimentally on a normal plot and a log-log plot for easy comparison against Fig. 4.8. $U_{pu,calculated}$ and $U_{pu,measured}$ agrees to within $\pm 15\%$, which is again reasonable. Comparing Figs. 4.8 and 4.9, the latter shows a slightly greater data-scatter than the former, which can be attributed to the former involving larger absolute values than the latter. Specifically, the dimensional U_{pu} values in Fig. 4.9 are one to two orders-of-magnitude smaller than the non-dimensional Re_p^* values. Since, the only error-inducing term is U_{pu} , Fig. 4.8, which deals with much larger absolute values than Fig. 4.9, naturally has a lesser data scatter.

This agrees with Kalman et al. (2005), where too, the plots involving non-dimensional entities (Re_p^* and Ar) had lower data scatter than those with dimensional ones (U_{pu} and d_p). Overall, the proposed pickup velocity model, based on the balance of drag and frictional forces, appears to be adequate in predicting the U_{pu} of all three types of binary mixtures of Zone I (or Geldart group B) particles. The model can thus provide a mechanistic insight into U_{pu} .

Fig. 4.9. Calculated versus measured pickup velocities: (a) normal axes and (b) log-log axes. The binary mixture properties used for the calculated U_{pu} values were obtained from Eqs. (4.18) to (4.20).

Fig. 4.10 shows a parity plot of $U_{pu,calculated}$ (Eq. (4.17)) versus $U_{pu,measured}$ values of Zone I particles obtained from literature (Cabrejos and Klinzing, 1992; Gomes and Mesquita, 2013; Goy et al., 2011; Kalman et al., 2005). Datasets where ϕ values were not reported are not included. Fig. 4.10 shows a reasonable agreement between $U_{pu,calculated}$ and $U_{pu,measured}$ within $\pm 25\%$. Hence, the proposed pickup velocity model can be used to predict U_{pu} values of spherical and non-spherical monodisperse and binary systems.

Fig. 4.10. Calculated ($U_{pu,calculated}$) versus measured ($U_{pu,measured}$) pickup velocities for data reported in literature.

4.4 Conclusion

Experiments were conducted to study the minimum pickup velocity (U_{pu}) of binary mixtures containing particles of different diameter (d_p), density (ρ_p) and sphericity (ϕ). Three types of binary mixtures were blended from glass, aluminum oxide and plastic particles. These

were binary-size, a binary-density and a binary-size-density mixtures. A force balance analysis was also carried out to theoretically determine U_{pu} .

It was found that (i) in a binary-size mixture, the U_{pu} of the mixture tended to mimic the U_{pu} of the larger component, and the mixture with the least spherical, larger component exhibited the greatest non-linearity in a U_{pu} versus composition plot; (ii) in a binary-density mixture, the U_{pu} of the mixture tended to mimic the U_{pu} of the denser component, especially at larger volume fractions of the denser species; (iii) in a binary-size-density mixture, if the larger component is denser, the U_{pu} of the mixture tended to mimic the U_{pu} of the larger and denser component; but if the smaller component is denser, the U_{pu} of the mixture either mimicked the U_{pu} of the larger component or none at all; (iv) shape effects had a non-negligible impact on the U_{pu} of Geldart group B binary mixtures, especially binary-size mixtures and binary-size-density mixtures when the smaller component was denser; (v) U_{pu} values of all binary mixtures agreed well with the Zone I pickup correlation (Kalman et al., 2005), and (vi) measured U_{pu} values agreed well with U_{pu} values calculated by a U_{pu} correlation derived from the drag and frictional force balance analysis.

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5

Minimum pickup velocity of nanoparticles

This chapter reports the U_{pu} values of nanoparticles. The differences between behaviors of nano-scale and micro-scale particles and polar and apolar nanoparticles in terms of U_{pu} are noted.

5.1 Experimental protocol

5.1.1 Experimental setup

The experimental setup schematically depicted in Fig. 5.1 was identical to that used in previous studies (Chapters 3 and 4). The pneumatic conveying setup was a hollow acrylic tube, and consisted of three sections, namely, A, B and C, which could be connected by means of screws and flanges. The inner diameter was 16 mm internal diameter and the wall thickness was 2 mm. Section A, the inlet section, was connected to the central air supply with a maximum flowrate of 0.0015 m³/s. Section B, the sample section, consisted of two semi-cylindrical halves, the bottom half of which was packed to the brim with plasticine up to a length of 27.5 cm from the inlet side, while the remaining 2.5 cm served as a chamber to hold the sample under investigation. More specifically, the surface of the particle sample was flat and of the same level as the plasticine to ensure that the cross-sectional area of airflow remained constant throughout Section B. This enabled the pickup velocity (U_{pu}) to be measured to a greater degree of accuracy compared to the method involving a varying cross-sectional area (Cabrejos and Klinzing, 1992, 1994; Hayden et al., 2003). The lengths of the inlet (Section A) and exit (Section C) sections were made sufficiently long to ensure a fully developed airflow before the particle sample and minimize exit effects, respectively. The outlet air stream was passed through water first and then through a High Efficiency Particulate Arrestance (HEPA) filter to

avoid the release of the nanoparticles into the environment (Tahmasebpour et al., 2013). Experiments were carried out at room temperature (approximately 25 °C). The relative humidity values in the laboratory and in the inlet gas stream were monitored to be $56 \pm 2\%$ and $27 \pm 2\%$, respectively, throughout the experiments. Regarding electrostatics, Valverde et al. (2008) reported that the electrostatic force (~ 0.01 nN) between nanoparticle agglomerates (silica, 200 μm) was about three orders-of-magnitude smaller than the van der Waal's force (~ 10 nN). Hence, electrostatic effects, negligible compared to the Van der Waals forces (Tahmasebpour et al., 2013; Valverde et al., 2008), were not considered in the present study.

Fig. 5.1. Experimental setup for measuring the minimum pickup velocity of nanoparticles

5.1.2 Materials

Nanoparticles in the range of 13 nm to 21 nm were investigated. The powders were polar and apolar silicon dioxide (SiO_2), polar and apolar titanium dioxide (TiO_2), and polar and apolar aluminum oxide (Al_2O_3). The polar nanoparticles had hydrophilic surfaces, while the apolar ones had hydrophobic surfaces. The latter were made so by substituting the hydroxyl groups of the polar nanoparticles by organic groups in a process called hydrophobization (Erdem et al., 2001; Schilde et al., 2010; Tahmasebpour et al., 2013). Table 5.1 lists the properties (namely, material, surface type, minimum fluidization velocity (U_{mf}), particle diameter (d_p), particle density (ρ_p) and Hamaker coefficient (A_H)) of the six nanoparticles

investigated in this work. These were the same nanoparticles investigated in a previous work on U_{mf} (Tahmasebpour et al., 2013), chosen with an objective to compare their fluidization and pickup behaviors, and the corresponding U_{mf} and U_{pu} values (Fig. 5.4 in Section 5.3). Note that no change in d_p was reported after hydrophobization, such that both the polar and apolar species of each material had approximately the same d_p values (Tahmasebpour et al., 2013).

Table 5.1. Properties of nanoparticles (Tahmasebpour et al., 2013)

Commercial name	Material	Surface type	U_{mf} (m/s)	d_p (nm)	ρ_p (kg/m ³)	A_H (J)
Aerosil 130	SiO ₂	Polar	0.042	16	2200	$6.6 \cdot 10^{-20}$
Aerosil R972		Apolar	0.006			
Aeroxide Alu C	Al ₂ O ₃	Polar	0.040	13	3600	$1.45 \cdot 10^{-19}$
Aeroxide Alu C805		Apolar	0.018			
Aeroxide P25	TiO ₂	Polar	0.050	21	4000	$1.54 \cdot 10^{-19}$
Aeroxide T805		Apolar	0.040			

5.1.3 Procedure to measure minimum pickup velocity

The pickup velocity, U_{pu} , is defined as the minimum air velocity at which mass loss due to entrainment becomes non-zero. The ‘weight loss’ method developed by Kalman et al. (2005) was adopted to determine U_{pu} . Apart from Fig. 5.1, the reader is referred to Fig. 3.1a for a detailed schematic of the experimental setup. Firstly, the empty section of the bottom semi-cylindrical half of Section B was loaded with the particle sample to be investigated and weighed with Mettler Toledo balance (ME204) which has a four decimal-place precision in grams. Care was taken to ensure that the surface of the particle bed was flat and of the same height as the plasticine mold. Secondly, the various sections of the setup (Fig. 5.1) were connected securely to ensure air-tightness. Thirdly, the airflow at the desired flow rate was commenced for a

duration of 120 s, which was the duration beyond which there was negligible weight loss per also determined in previous work (Goy et al., 2011; Tay et al., 2012). Fourthly, at the end of 120 s, the air supply was shut off, and the bottom semi-cylindrical half of Section B was weighed to ascertain the mass loss of the particle sample. This procedure was repeated at decreasing rates of airflow till the mass loss approaches zero. At least three readings at each airflow velocity were taken to check for reproducibility.

5.2 Analysis of mass loss curves

Fig. 5.2 presents the mass loss curves (i.e., mass loss versus air velocity) obtained for all six samples. Notably, the point on the x -axis where the extrapolated mass loss curve intersects the x -axis was determined as the U_{pu} of the nanoparticle species (Anantharaman et al., 2015; Goy et al., 2011). This was unlike the determination of U_{pu} for the micron-scale particles in Chapters 3 and 4, where the gas velocity at a threshold of 0.01 g mass loss was considered as the U_{pu} . An extrapolation method was applied here instead, since the mass loss for nanoparticles near pickup was far lesser than 0.01 g and a new lower threshold could not be set because the lowest non-zero mass loss values exhibited by different species varied by a factor of up to four. The U_{pu} values of the six nanoparticle species are displayed in Fig. 5.3 (with error bars representing two standard deviations), which clearly shows that polar species have higher U_{pu} values than the apolar ones.

Fig. 5.2 Mass loss curves

Fig. 5.3 Minimum pickup velocity values of apolar and polar nanoparticles. The filled markers represent apolar particles while the unfilled markers represent polar particles.

Three observations on the differences between the apolar and polar species exhibited in the mass loss curves (Fig. 5.2) and U_{pu} values (Fig. 5.3) are worth highlighting: (i) for each nanoparticle species, the apolar mass loss trends lie above the polar ones, which suggest the lesser extent of agglomeration for the former, (ii) the differential between the polar and apolar mass loss curves and U_{pu} values decreased in the order of SiO_2 , Al_2O_3 , and TiO_2 , and (iii) the apolar species assumed a more S-shaped profile than the polar ones, with apolar SiO_2 having the most pronounced S-shaped curve. The distinctly different behaviors observed between apolar and polar nanoparticles of the three materials are due to the nature of the inter-particle

interactions stemming from the different surface properties. On one hand, the polar particles have hydroxyl groups on the surface, and hence, hydrogen bonds form readily between the particles. On the other hand, because apolar particles are such that the hydroxyl groups have been replaced by organic groups via hydrophobization, inter-particle hydrogen bonds are absent, thereby the inter-particle cohesion is comparatively weaker (Tahmasebpour et al., 2013). Therefore, with respect to (i), as Fig. 5.2 shows for each nanoparticle type, the mass loss of the apolar species was greater than the polar ones due to the lesser extents of agglomeration in the former. Correspondingly, the mass loss curves of apolar nanoparticles intersect the x -axis at smaller values, thus giving lower U_{pu} values than the polar nanoparticles.

Regarding (ii), the strength of the inter-particle interaction also underlies the observation that polar and apolar SiO_2 nanoparticles exhibited the largest difference in mass loss and U_{pu} , while TiO_2 nanoparticles the least. Table 5.1 displays the Hamaker (1937) coefficients, which indicate the strength of inter-particle cohesion, and are distinct from the polarity-based hydrogen bonding. The Hamaker coefficient of SiO_2 is the smallest among the three nanoparticle species (Table 5.1). This implies that it has the least tendency for inter-particle cohesion for the apolar species. Hence, the presence of hydrogen bonds in the polar species represents a significant increase of inter-particle cohesion, thus resulting in the greatest difference in the mass loss curves and U_{pu} values. On the other hand, although TiO_2 has a higher Hamaker coefficient than Al_2O_3 , the largest particle diameter among the three nanoparticle species implies the least diameter-based van der Waals attraction. Consequently, the interplay of the twin effects represented by the Hamaker coefficient and hydrogen bonding results in the lowest differential between the apolar and polar species of TiO_2 .

As for (iii), the most distinctly S-shaped mass loss curve of apolar SiO_2 is due to the low tendency for inter-particle cohesion (in terms of lowest Hamaker coefficient and lack of hydrogen bonding) among all six species. As with gas–solid fluidization too, SiO_2 tends to

exhibit ‘agglomerate particulate fluidization’ as opposed to ‘agglomerate bubbling fluidization’ per other nanoparticles (Tahmasebpour et al., 2013; Yao et al., 2002). Specifically, the low inter-particle cohesion of SiO₂ nanoparticles implies they tend to form smaller agglomerates, which were more easily picked up at lower air velocity, and then the steeper increase in mass as velocity increased was due to the larger differential between the air velocity and U_{pu} (Fig. 5.2).

Comparing the U_{pu} values obtained here (Fig. 5.3) to the minimum fluidization velocities (U_{mf}) reported of the same nanoparticles (listed in Table 5.1) (Tahmasebpour et al., 2013), Fig. 5.4 shows the ratio $\frac{U_{mf,polar}/U_{mf,apolar}}{U_{pu,polar}/U_{pu,apolar}}$ versus the three species (namely, SiO₂, Al₂O₃ and TiO₂), where $U_{mf,polar}$ and $U_{mf,apolar}$ are the minimum fluidization velocities of the polar and apolar variants of a species, respectively, and $U_{pu,polar}$ and $U_{pu,apolar}$ are the minimum pickup velocities of the polar and apolar variants of the same species, respectively. Fig. 5.4 indicates that $U_{mf,polar}/U_{mf,apolar}$ is approximately between 1 and 3.5 times that of $U_{pu,polar}/U_{pu,apolar}$. This is presumably due to the fact that extents of hydrogen bonding experienced by the polar nanoparticles in a fluidized bed are greater than that involved in pickup in a horizontal pneumatic conveying system. Specifically, the fluidized nanoparticles are largely surrounded on all sides by particles, whereas the nanoparticles involved in the determination of U_{pu} are those at the surface of the particle bed. Hence, the number of adjacent contacts for hydrogen bonding is a lot greater in the fluidized bed than that for the horizontal pneumatic conveying system. The ratio is the largest for SiO₂ and smallest for TiO₂ presumably due to the aforementioned effect, namely, the former species having the lowest Hamaker coefficient and the latter the greatest, being amplified for SiO₂ more than TiO₂ in the fluidized bed due to the greater number of adjacent contacts than in the horizontal pneumatic conveying system.

Fig. 5.4 $\frac{U_{mf,polar}/U_{mf,apolar}}{U_{pu,polar}/U_{pu,apolar}}$ for the three nanoparticle species

Two effects with regard to the agglomeration behavior are acknowledged but detailed investigations are beyond the scope of the current effort: (i) agglomerates were already present in the nanoparticle samples even before subjecting them to entrainment, as ‘stored nanoparticle agglomerates’ have been reported to be formed during storage, transport, and processing due to shock and vibrations (de Martin et al., 2014), and (ii) a critical particle concentration likely exists for the onset of agglomeration during entrainment. Notably, the current effort focuses only on the agglomerates observed during pickup, which may result from agglomeration during storage and/or entrainment, as is the case in practical applications.

5.3 Comparison against the Three-Zone model

Notably, Fig. 5.3 shows that the U_{pu} values are an order-of-magnitude lower than that for micron-sized particles (Anantharaman et al., 2015; Cabrejos and Klinzing, 1992, 1994; Gomes and Mesquita, 2013; Goy et al., 2011; Hayden et al., 2003; Kalman et al., 2005; Tay et al., 2012) or that predicted by the U_{pu} master curve (Kalman et al., 2005), which is unexpected from earlier reports stating that U_{pu} increases as d_p decreases (Cabrejos and Klinzing, 1992; Gomes and Mesquita, 2013; Hayden et al., 2003) for Zone III (Kalman et al., 2005) (or Geldart Group C (Geldart, 1973)), wherein the nanoparticles belong. Clearly, Fig. 5.3 reveals that the previously observed U_{pu} trends for micron-sized particles do not hold for nanoparticles, which is further affirmed in the master curve (i.e., a plot of Re_p^* versus Ar , whereby Re_p^* is the particle Reynolds number modified to account for different pipe diameters and Ar is Archimedes number) for U_{pu} reported by Kalman et al. (2005). Fig. 5.5 shows the Re_p^* versus Ar plot, acknowledged for designating the three zones distinguishing three different behaviors in pneumatic conveying systems. The reader is requested to refer to Chapter 2 for more details on the Three-Zone model. The expressions for Re_p^* and Ar , and the empirical correlations for the pickup Zones are reiterated here for convenience.

$$Re_p^* = \frac{\rho_p d_p U_{pu}}{\mu_f \left[1.4 - 0.8 \exp\left(-\frac{D/D_{ref}}{1.5}\right) \right]} \quad (2.5)$$

$$Ar = \frac{g \rho_f (\rho_p - \rho_f) d_p^3}{\mu_f^2} \quad (2.6)$$

where μ_f is the ambient air viscosity, D is the pipe diameter used, D_{ref} is the reference pipe diameter of 50 mm (Goy et al., 2011; Kalman et al., 2005; Tay et al., 2012), g is gravitational acceleration, and ρ_f is the density of air. The empirical correlations of the Three-Zone model are (Kalman et al., 2005)

$$\text{Zone I: } Re_p^* = 5Ar^{\frac{3}{7}} \quad \text{for } Ar \geq 16.5 \quad (2.2)$$

$$\text{Zone II: } Re_p^* = 16.7 \quad \text{for } 0.45 < Ar < 16.5 \quad (2.3)$$

$$\text{Zone III: } Re_p^* = 21.8Ar^{\frac{1}{3}} \quad \text{for } Ar \leq 0.45 \quad (2.4)$$

Recall that Zones I, II and III approximately corresponds to Geldart Groups B, A and C (Geldart, 1973), respectively.

Fig. 5.5 Re_p^* versus Ar plot (using particle properties, namely, d_p and ρ_p , listed in Table 5.1). The solid line represents the correlations for each Zone, the dotted line represents the extrapolated Zone I correlation, while each 'x' represents each of the six nanoparticles investigated.

Surprisingly, Fig. 5.5 shows that all six nanoparticles (represented by 'x' in Fig. 5.5), which should fall into Zone III by virtue of the Re_p^* and Ar values, had Re_p^* values at least an order-of-magnitude smaller than that predicted by the Zone III correlation. Instead, the nanoparticles

agreed with the extrapolated Zone I correlation, which indicate that the nanoparticles behaved more like Zone I (or Geldart group B) particles.

Fig. 5.6 High-speed camera images at 2 ms intervals showing polar Al_2O_3 nanoparticles being picked up in agglomerates. The air flow direction is from right to left in all frames.

High-speed camera imaging of the nature of the pick-up mechanism reveals that, unlike their micron-sized counterparts, the nanoparticles were consistently entrained in lumps or agglomerates, as shown in Fig. 5.6. It should be noted that the length of Section C and consequently the downstream conveying time have little effect on the pickup of nanoparticles as agglomerates. The agglomeration phenomenon implies that the actual ρ_p and d_p values are lower and higher, respectively, than that given for the individual nanoparticles (Yao et al., 2002)

(Table 5.1), which explains the deviation from the predictions by the available correlation (Kalman et al., 2005).

Note that agglomerates are already present in the nanoparticle samples before subjecting them to entrainment. The smallest nanoparticle aggregates are formed by sintering in the flame reactor, and their inter-particle bonds are very strong due to their covalent nature. These sintered aggregates further link to form larger and softer agglomerates. They are referred to as ‘stored nanoparticle agglomerates’ and are formed during storage, transport and processing due to shock and vibrations (de Martin et al., 2014), and are present in the nanoparticle samples to be tested. According to (de Martin et al., 2014), nanoparticle agglomerates larger than the fragmentation scale ($\sim 10\ \mu\text{m}$) are weak and have low relative strength. Stored nanoparticle agglomerates are larger than this threshold and are subject to fragmentation and re-agglomeration. Hence, like fluidization, this phenomenon is expected to occur at incipient pneumatic conveying, albeit to a lesser extent than the former.

To predict the particle diameter (d^{**}) and density (ρ^{**}) of fluidized nanoparticle agglomerates, models based on force balance have been put forth. In particular, de Martín and van Ommen (2013) developed a model to calculate the size of complex nanoparticle agglomerates based on the attractive forces between micron-sized primary agglomerates (with particle diameter, d^* , and density, ρ^*), which form the constituent building blocks for the final complex agglomerates. The factoring in of nanoparticle diameter (d_p) as asperities on the agglomerate surface led to weaker inter-agglomerate forces for larger nanoparticles, which resulted in more accurate predictions of agglomerate sizes in dry fluidized beds (de Martín and van Ommen, 2013). This model was adopted to estimate the properties of the primary and complex nanoparticle agglomerates in this work, as displayed in Table 5.2 for all six nanoparticles. A similar force balance is used to estimate the diameter and density of

agglomerates in the present work. In a two level hierarchical fractal consisting of primary and complex agglomerates,

$$\frac{\rho^*}{\rho_p} = k_0 \left(\frac{d^*}{d_p} \right)^{D_0-3} \quad (5.1)$$

$$\frac{\rho^{**}}{\rho^*} = k_1 \left(\frac{d^{**}}{d^*} \right)^{D_1-3} \quad (5.2)$$

where ρ^* , d^* , D_0 and ρ^{**} , d^{**} , D_1 are the densities, diameters and fractal dimensions of primary and complex agglomerates respectively. k_0 and k_1 are equal to unity (Nam et al., 2004). D_0 and D_1 are considered 2.6 (Wang et al., 2006) and 2.0 (de Martín et al., 2014) respectively. The Van der Waal force of attraction between two primary agglomerates is given by

$$F_{vdw} = \frac{A_H d^* \left(\frac{\rho^*}{\rho_p} \right)^2}{24 z_{eq}^2} \quad (5.3)$$

where A_H is the Hamaker coefficient and z_{eq} is the equivalent distance between two agglomerates, which can be considered as the distance between two smooth spheres that creates the same attractive force if those spheres had asperities. The A_H values for the three materials are given in Table 5.1. A value of 35 μm is considered for d^* for all six nanopowders and z_{eq} is defined by (de Martín and van Ommen, 2013)

$$z_{eq} = (0.4 + 0.063 d_p) \cdot 10^{-9} \quad (5.4)$$

As explained above, interparticle forces due to hydrogen bonds are non-negligible in polar nanoparticles and have to be added to the total binding forces in agglomerates. The force of attraction due to hydrogen bonds between two primary agglomerates is given by (de Martín and van Ommen, 2013)

$$F_{OH} = \alpha d^* \quad (5.5)$$

where, α is a parameter proportional to strength of the hydrogen bond, concentration of hydroxyl groups on the agglomerate surface, fraction of agglomerate surface exposed to the bonding and the difference between the maximum range of H-bond interaction and minimum interparticle distance. The sum of F_{vdw} (Eq. (5.3)) and F_{OH} (Eq. (5.5)) is the total attractive force on the agglomerate. It is equated to the total drag on the agglomerate, which is equal to the normal force, along the same lines of the equilibrium criterion employed by (de Martín and van Ommen, 2013)

$$N \left(\frac{A_H d^* \left(\frac{\rho^*}{\rho_p} \right)^2}{24 z_{eq}^2} + \alpha d^* \right) = \frac{\pi}{6} g d^{**3} \left(\rho^* \left(\frac{d^{**}}{d^*} \right)^{-1} - \rho \right) \quad (5.6)$$

N is a fitting parameter to include the connectivity of the primary agglomerates. It is associated with the number of links between primary agglomerates that are required to break a complex agglomerate (de Martín and van Ommen, 2013). ρ is the density of air (1.2 kg/m³). Table 5.2 lists the parameters in Eq. (5.6) and their values:

Table 5.2. Parameter values (de Martín and van Ommen, 2013) from Eq. (5.6)

Parameter (unit)	Value
N (-)	15.08
z_{eq} (m)	$(0.4 + 0.063 * 10^9 d_p) * 10^{-9}$
α (N/m)	$2.5 * 10^{-6}$
g (m/s ²)	9.81

Hence, the complex agglomerate diameter is obtained by solving Eq. (5.6) for d^{**} . Then ρ^{**} is obtained from Eq. (5.2). Readers interested in more details are referred to (de

Martín and van Ommen, 2013), wherein the typical parameters reported are similarly used here. Two notes are worth highlighting: (i) Table 5.3 shows clearly that the complex nanoparticle agglomerates had particle diameters four orders-of-magnitude greater than and particle densities two orders-of-magnitude lesser than the individual nanoparticles (Table 5.1), and (ii) model (de Martín and van Ommen, 2013) predictions and experimental values agreed well in that while the polar Al_2O_3 nanoparticle agglomerates ranged in diameters from 0.1 to 0.8 mm in Fig. 5.6, the model predicts that the complex nanoparticle agglomerates of this species have a characteristic diameter (d^{**}) of 0.46 mm in Table 5.3. It was interesting to note that in experiments carried out over multiple days, nanoparticles of all species entrained as agglomerates in the same size range.

Table 5.3. Primary (Nam et al., 2004; Valverde and Castellanos, 2008) and Complex (de Martín and van Ommen, 2013) nanoparticle agglomerate properties

Nanoparticle	Primary agglomerate diameter, d^* (μm)	Primary agglomerate density, ρ^* (kg/m^3)	Complex agglomerate diameter, d^{**} (μm)	Complex agglomerate density, ρ^{**} (kg/m^3)
SiO_2 (Polar)	35.0	101.5	429.5	8.3
SiO_2 (Apolar)	35.0	101.5	308.8	11.5
Al_2O_3 (Polar)	35.0	152.8	458.7	11.7
Al_2O_3 (Apolar)	35.0	152.8	392.9	13.6
TiO_2 (Polar)	35.0	205.7	353.0	20.4
TiO_2 (Apolar)	35.0	205.7	292.9	24.6

Fig. 5.7 shows the values of the primary and complex nanoparticle agglomerates on the master curve of Re_p^* versus Ar (Kalman et al., 2005). The nanoparticles, primary agglomerates and complex agglomerates are different embodiments of the same species, hence U_{pu} values used for calculating Re_p^* are the same (Fig. 5.3). The diameter values, however, are those of the entities for which the dimensionless numbers are calculated i.e. the particle diameter values used for calculation of Re_p^* and Ar of the nanoparticles, primary agglomerates and complex agglomerates are d_p (Table 5.1), d^* (Table 5.3) and d^{**} (Table 5.3), respectively. Despite nanoparticles picking up as micron-sized agglomerates, and not as individual particles, d_p , the individual particle diameter was used to compare U_{pu} results against the Three Zone Model (Figs. 5.5 and 5.7) because in terms of particle size, d_p is the inherent, defining and unchanging property of a species. Actual d^* and d^{**} values vary from agglomerate to agglomerate (Fig. 5.6) and an uncertainty of $\pm 30\%$ in the calculation of d^{**} by the model is acknowledged (de Martín and van Ommen, 2013). Hence, a nanoparticle species undergoing pickup is best represented by d_p .

It can be seen from Fig. 5.7 that, interestingly, the individual nanoparticles, and the primary and complex nanoparticle agglomerates are all well-represented by the Zone I correlation. This underscores the observation that, although nanoparticles fall in Zone III (Kalman et al., 2005) (or Geldart group C (Geldart, 1973)), they behave like Zone I (Kalman et al., 2005) (or Geldart group B (Geldart, 1973)) particles in pneumatic conveying systems. Hence, despite the change in particle diameter due to agglomeration, nanoparticles continued to adhere to Zone I, showing the dominance of U_{pu} (the unchanged parameter) in dictating Zones. Interestingly, for both the cases of primary and complex agglomerates, apolar SiO_2 nanoparticles exhibit the largest deviations from the correlations. Recall that SiO_2 also gave the largest differential between the polar and apolar species in the mass loss curves (Fig. 5.2), U_{pu} values (Fig. 5.3), and also U_{mf} values (Tahmasebpour et al., 2013). Therefore, SiO_2 makes

an interesting future case study on the interplay of hydrogen bonding and Hamaker attractions in nanoparticle agglomeration behavior.

Fig. 5.7. Re_p^* versus Ar : Nanoparticles, and primary and complex agglomerates of each nanoparticle species. The solid line represents the correlations for each Zone, the dotted line represents the extrapolated Zone I correlation, while each discrete data point represents each of the six nanoparticles investigated.

5.4 Conclusion

Experiments involving horizontal gas–solid pneumatic conveying have been carried out for six nanoparticle species of different materials (i.e., SiO_2 , Al_2O_3 , and TiO_2) and surfaces (i.e., apolar and polar). The mass loss curves (i.e., mass loss vs. velocity) were generated to determine the U_{pu} values via the weight loss method (Kalman et al., 2005). Considering that

nanoparticles were picked up as agglomerates rather than individually, the predicted diameter and density values of the primary and complex agglomerates were calculated (de Martín and van Ommen, 2013). Using the experimentally obtained U_{pu} values, the Ar and Re_p^* for the individual nanoparticles, and primary and complex agglomerates were compared against the correlations available (Kalman et al., 2005).

Results indicate that (1) compared to polar nanoparticles which had additional inter-particle hydrogen bonding, apolar ones displayed higher mass loss values at the same velocities and lower U_{pu} values, (2) due to the interplay of the twin effects represented by diameter-based Hamaker and surface-based hydrogen bonding, the greatest discrepancy between apolar and polar surfaces was exhibited by SiO_2 , followed by Al_2O_3 then TiO_2 , (3) the difference between polar and apolar species was greater in the case of U_{mf} than for U_{pu} , due to the increased extents of hydrogen bonding in the former case of fluidized nanoparticles largely surrounded on all sides by particles, than in the latter case whereby the nanoparticles involved in the determination of U_{pu} are those at the surface of the particle bed, (4) U_{pu} values were at least an order-of-magnitude lower than that expected from the well-acknowledged U_{pu} correlation (Kalman et al., 2005), because the nanoparticles were picked up as porous, micron-sized agglomerates (de Martín and van Ommen, 2013) rather than individual nanoparticles, (5) although individual nanoparticles fall in Zone III (Kalman et al., 2005) (or Geldart group C), their behavior corresponded more with Zone I (Kalman et al., 2005) (or Geldart group B) particles in pneumatic conveying systems.

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6

Minimum pickup velocity – the transition between the nano-scale and the micro-scale

This chapter attempts to reveal the missing link between nano-scale and micro-scale particles in terms of U_{pu} . Modifications to the Three-Zone model are also proposed.

6.1 Experimental protocol

The experimental unit and the method to determine U_{pu} used here are identical that used in the previous nanoparticles study (Chapter 5). The reader is referred to Sections 5.1.1 and 5.1.3 for details on the experimental unit and the procedure to measure U_{pu} respectively.

6.1.1 Materials

Alumina (Al_2O_3) particles in the particle diameter (d_p) range of 5 to 300 nm were obtained from US-Nano, and with the particle diameters (d_p) of 20,000 and 110,000 nm were obtained from Sasol. Alumina (Al_2O_3) particles have a particle density (ρ_p) of 3600 kg/m^3 . The polymorph types (i.e., gamma or alpha) and particle diameters (d_p) of the nine samples are listed in Table 6.1. The particles were used as is. It should be noted that the particle diameters (d_p) were provided by the vendors, who did not provide the particle-size distributions (PSDs), although it has been reported that changes in PSDs affect the pickup velocity (U_{pu}) for micro-scale particles (Chapter 3). Efforts to obtain the PSDs for the nanoparticles via a nano-sizer (BIC 90Plus) were futile since larger mean particle diameters (d_p) than those provided by the vendor were obtained presumably due to insufficient dispersion of the nanoparticle agglomerates.

Table 6.1. Relevant properties of the alumina (Al₂O₃) particles investigated

d_p (nm)	Polymorph type
5	Gamma
20	Gamma
80	Gamma
80	Alpha
135	Alpha
200	Alpha
300	Alpha
20,000	Gamma
110,000	Gamma

6.2 Bridging the gap between the nano-scale and micro-scale

To enable a fair comparison across the various studies, Kalman et al. (2005) proposed a correlation to normalize the effect of different diameters of the pneumatic conveying channels used with respect to that of a channel diameter of 50 mm:

$$\frac{U_{pu}}{U_{pu,50}} = 1.4 - 0.8 \cdot e^{-\frac{D/D_{50}}{1.5}} \quad (6.1)$$

where U_{pu} is the minimum pickup velocity, $U_{pu,50}$ is the minimum pickup velocity normalized for a channel inner diameter of 50 mm, D is the inner diameter of the pneumatic conveying channel and D_{50} is the reference channel diameter of 50 mm. Note that Eq. (2.5) for the particle Reynolds number, Re_p^* , is derived from Eq. (6.1) (Kalman et al., 2005).

Fig. 6.1 displays minimum pickup velocity (U_{pu}) and normalized minimum pickup velocity ($U_{pu,50}$; Eq. (1)) versus particle diameter (d_p), specifically the individual particle diameters (d_p) listed in Table 6.1, on a semi-logarithmic plot of all the particle samples investigated in this study. Data from four previous studies are also portrayed: three of them were included because alumina particles were similarly studied (Anantharaman et al., 2015; Cabrejos and Klinzing, 1992; Hayden et al., 2003), while one was on zirconium particles ($\rho_p = 5964 \text{ kg/m}^3$) because it represented the uppermost limit of Ar investigated to date (Kalman et al., 2005). Four short notes on the data obtained in this study depicted in Fig. 6.1 include (i) the trends for U_{pu} (Fig. 6.1a) and $U_{pu,50}$ (Fig. 6.1b) are very similar, which implies that the normalization with respect to pipe diameter (Eq. (6.1)) does not significantly affect the overall trends; (ii) the U_{pu} magnitudes for the particles with $d_p = 5\text{-}135 \text{ nm}$ were an order-of-magnitude lower than that for micro-scale particles investigated in previous efforts (Cabrejos and Klinzing, 1992; Hayden et al., 2003; Kalman et al., 2005), which is consistent with the data for the nano-scale particles with $d_p = 13\text{--}21 \text{ nm}$ obtained by our previous study (Anantharaman et al., 2015); (iii) the U_{pu} characterized for the alumina particles with $d_p = 20,000 \text{ nm}$ and $110,000 \text{ nm}$ agreed qualitatively with the data obtained by previous studies (Cabrejos and Klinzing, 1992; Hayden et al., 2003); and (iv) the difference between the U_{pu} values of the alpha and gamma variants of the alumina particles with $d_p = 80 \text{ nm}$ (Table 6.1) was not significant, hence the effect of polymorph type can be considered as negligible.

Fig. 6.1. Comparison of experimental results from this study (the d_p values on the x -axis are of the individual particles listed in Table 6.1) with those from previous studies (Cabrejos and Klinzing, 1992; Hayden et al., 2003; Kalman et al., 2005) and Chapter 5: (a) minimum pickup velocity (U_{pu}), and (b) normalized minimum pickup velocity ($U_{pu,50}$; Eq. (6.1)).

The significant highlight in Fig. 6.1 is that the U_{pu} values exhibited a non-monotonic trend with respect to d_p . Specifically, U_{pu} increased slowly between $d_p = 5 - 80$ nm then quickly till $d_p = 530$ nm, followed by a plateau between $d_p = 530$ nm – 15,000 nm, after which a decrease between $d_p = 15,000$ nm – 110,000 nm before increasing beyond $d_p = 110,000$ nm. For the nano-scale particles, although they belong to Zone III (or Geldart Group C), it was found earlier (Chapter 5) that they surprisingly behaved more like Zone I (or Geldart Group B) particles due to agglomeration effects. On the other hand, for the micro-scale particles ($d_p \geq 530$ nm), a non-monotonic trend has been reported in that U_{pu} plateaued then decreased with d_p in Zones III and II (or Geldart Groups C and A, respectively) due to decreasing inter-particle cohesion effects, then increased with d_p in Zone I (or Geldart Group B) due to increasing inertial effects (Cabrejos and Klinzing, 1992; Hayden et al., 2003). Notably, Fig. 6.1 serves to reveal the missing link on the horizontal pneumatic conveying behavior of particles between $d_p = 5$ nm (i.e., nano-scale) and $d_p = 530$ nm (Hayden et al., 2003)(i.e., micro-scale) in terms of U_{pu} trends.

Fig. 6.2 illustrates the plot of Re_p^* (Eq. (2.5)) vs. Ar (Eq. (2.6)), whereby the d_p values substituted were those of the individual particles listed in Table 6.1. The solid lines represent the three correlations for each of the zones (Eqs. (2.2) – (2.4)), while the dotted line represents the extrapolated Zone I correlation (Eq. (2.2)). Each discrete data point represents each of the nine particle samples (Table 6.1). Three observations are notable from Fig. 6.2. First, the particles with $d_p = 20,000$ and $110,000$ nm agreed exactly with the Zone II and Zone I correlations, respectively, which is expected. For such micro-scale particles, the smaller ones in Zone I experience some inter-particle cohesion effects, while the larger ones tend to be entrained as individual particles (Cabrejos and Klinzing, 1992; Hayden et al., 2003; Kalman et al., 2005). Second, although nano-scale particles belong to Zone III, the particles with particle diameter (d_p) up to 135 nm agreed well with the extrapolated Zone I correlation. This is similar to the observations in the previous study (Chapter 5), which attributed it to agglomeration effects (de Martín and van Ommen, 2013) that leads to micro-scale agglomerates rather than nano-scale particles being entrained. Third and perhaps most notably, the particles with $d_p = 200$ and 300 nm agreed with none of the three zones. More specifically, these two particle samples lie in between the extrapolated Zone I and Zone III correlations, which indicated an intermediate mechanism between the pneumatic conveying of micro-scale agglomerates (Zone I) and smaller agglomerates (Zone III).

Fig. 6.2. Re_p^* vs Ar for the particle samples investigated. The solid lines represent the three correlations for each of the Zones and the dotted line represents the extrapolated Zone I correlation. Each discrete data point represents each of the nine particle samples. Re_p^* (Eq. (2.5)) and Ar (Eq. (2.6)) were calculated using the individual particle diameter (d_p) listed in Table 6.1.

To estimate the characteristic diameters (d^{**}) and densities (ρ^{**}) of the complex nanoparticle agglomerates, the force balance model presented in Chapter 5 was employed. This model was adopted from de Martín and van Ommen (2013), wherein the diameters of complex nanoparticle agglomerates formed during dry nano-scale particle fluidization was calculated based on inter-particle cohesive forces. Table 6.2 provides the properties (namely, particle diameter and particle density) of the primary and complex nanoparticle agglomerates. Specifically, nanoparticle agglomerates have been found to possess a hierarchical structure (de

Martín et al., 2014; Yao et al., 2002), as in that primary nanoparticle agglomerates (Castellanos et al., 2001; Nam et al., 2004; Quintanilla et al., 2012; Wang et al., 2006) of diameter d^* are first created which further agglomerate to form complex agglomerates (Castellanos et al., 2001; de Martín and van Ommen, 2013) of diameter d^{**} . It should be noted that the particles with $d_p = 20,000$ and $110,000$ nm were omitted from Table 6.2, because the agglomerate diameters so obtained were smaller than the individual particle diameter, which is not physical. Three noteworthy observations are obtained from Table 6.2. Firstly, the diameters of the complex agglomerates (d^{**}) were of the order 10^5 nm, which is three to five orders-of-magnitude greater than the individual particle diameter (Table 6.1), while the complex agglomerate density (ρ^{**}) were 10^1 - 10^2 kg/m³, which is one to three orders-of-magnitude smaller than the individual particle density. This is consistent with the results of Chapter 5. Secondly, as the individual particle diameter (d_p) increased, the complex agglomerate diameter (d^{**}) decreased while the complex agglomerate density (ρ^{**}) increased, which indicates the decreased inter-particle cohesion effect and the decreased porosity of the resulting agglomerates. Thirdly, the ρ^{**} value corresponding to the particle diameter (d_p) of 5 nm particles appears to be low at 5.5 kg/m³, which translates to an unexpectedly high particle porosity of 0.999. This is perhaps not as surprising considering: (i) the ρ^{**} and porosity values of particles with $d_p = 12$ -13 nm have been reported to be 13 kg/m³ (de Martín et al., 2014; Wang et al., 2006; Wang et al., 2007) and 0.997 (Wang et al., 2007), respectively; and (ii) an error of up to $\pm 30\%$ in the prediction of d^{**} by the model has been acknowledged (de Martín and van Ommen, 2013), and since the model was developed only for particle diameter, the values for density and porosity may be subject to even greater errors.

Table 6.2. Properties of primary (Nam et al., 2004; Valverde and Castellanos, 2008) and complex (de Martín and van Ommen, 2013) agglomerates of nano-scale particles.

d_p (nm)	Primary agglomerate diameter, d^* (nm)	Primary agglomerate density, ρ^* (kg/m ³)	Complex agglomerate diameter, d^{**} (nm)	Complex agglomerate density, ρ^{**} (kg/m ³)
5	35,000	104.29	669,500	5.45
20	35,000	181.59	374,900	16.95
80	35,000	316.16	196,600	56.28
80	35,000	316.16	196,600	56.28
135	35,000	389.77	161,000	84.73
200	35,000	456.13	141,400	112.90
300	35,000	536.44	125,600	149.49
Note that the particles with $d_p = 20,000$ nm and 110,000 nm are excluded as unphysical d^{**} values were obtained.				

Regarding the nature of the entities (i.e., particles or agglomerates) being pneumatically conveyed, the gradient of the mass loss curve provides a good indication. Accordingly, the gradients of all the mass loss curves were computed by linear regression analyses of the five or six points constituting each curve. The linear regression coefficient (R^2) values were all greater than 0.94, which implied reasonable goodness of fit and hence that the mass loss curves were approximately straight lines for the five or six data points corresponding to air velocities (U) just above the minimum pickup velocity (U_{pu}). Fig. 6.3 shows an example of the mass loss curve of the 20 nm particles. The x -axis represents the air velocity (U) while the y -axis

represents mass loss. As can be seen from Fig. 6.3, the curve can be approximated as a straight line with a slope of 0.0661 g/(m/s) and regression coefficient $R^2 = 0.9832$.

Fig. 6.3. Linear approximation of the mass loss curve of 20 nm particles. The line has a slope of 0.0661 g/(m/s) and regression coefficient R^2 of 0.9832.

Fig. 6.4 shows the average slopes of the linearly regressed mass loss curves vs. particle diameter (d_p), specifically the individual particle diameters (d_p) listed in Table 6.1, on a semi-logarithmic plot. Each error bar indicates the span of values for the three repeats carried out. The average slope is approximately invariant with particle diameter (d_p) for $d_p \leq 135$ nm, then clearly decreases before increasing with d_p . The zones depicted in Fig. 6.2 can also be seen in Fig. 6.4, which also serves to provide a mechanistic understanding of the nature of the entities being entrained. Regarding Zone I, particles with $d_p \leq 135$ nm and $d_p = 110,000$ nm were classified into this zone (Fig. 6.2), which is such that the entrained particles or agglomerates are on the micro-scale (Table 6.2). In Fig. 6.4, Zone I is represented by larger slope magnitudes, which indicate the entrainment of larger entities beyond the minimum pickup velocity (U_{pu}).

With respect to Zone II, particles with $d_p = 20,000$ nm were categorized into this zone (Fig. 6.2), which is such that the entrained particles are not individual particles due to inter-particle cohesion, but the cohesion effects are not adequately significant for the agglomerates to be the size of the entities entrained in Zone I. In Fig. 6.4, Zone II is represented by the smallest slope magnitude. Finally, the particles with $d_p = 200$ nm and 300 nm agreed with neither of the three zones in Fig. 6.2 and are represented by slope magnitudes in between those for Zones I and II in Fig. 6.4.

Fig. 6.4. Slope of the linearly regressed mass loss curves versus particle diameter (d_p), specifically the individual particle diameters (d_p) listed in Table 6.1.

Fig. 6.5 shows the Re_p^* against Ar plot for the primary and complex agglomerates of the particles with particle diameters (d_p) of 5–300 nm (Table 6.2). In contrast to Fig. 6.2, the particle diameter (d_p) values used in the calculation of Re_p^* (Eq. (2.5)) and Ar (Eq. (2.6)) are those of the primary and complex agglomerates listed in Table 6.2. Fig. 6.2 has shown that

particles in the particle diameter (d_p) range of 5–135 nm agreed well with the extrapolated Zone I correlation, while the two particle samples with $d_p = 200$ and 300 nm did not agree with any of the Zone. Here, Fig. 6.5 indicates that, for particles with $d_p = 5$ –135 nm, the primary agglomerates agreed somewhat with the extrapolated Zone I correlation, while the complex agglomerates agreed with the Zone I correlation. This is consistent with the previous study for particle diameters (d_p) in the range of 13–21 nm (Chapter 5). In addition, both the primary and complex agglomerates of the particle samples with $d_p = 200$ and 300 nm persist in not agreeing well with any Zone.

Fig. 6.5. Re_p^* vs Ar plot of the primary agglomerate and complex agglomerates for the particles with $d_p = 5$ nm - 300 nm (Table 6.2). The solid lines represent correlations for the three pickup zones and the dotted line represents the extrapolated Zone I correlation. Re_p^* (Eq. (2.5)) and Ar (Eq. (2.6)) were calculated using the primary (d^*) or complex (d^{**}) nanoparticle diameters listed in Table 6.2.

6.3 Modifications to the Three-Zone model

To further assess the categorization of particles into Zones (Kalman et al., 2005) for the intermediate particle diameters, Fig. 6.6 summarizes the minimum pick-up velocity (U_{pu}) data to date, per those depicted in Fig. 6.2, in a plot of Re_p^* against Ar . Similar to Fig. 6.2, the d_p values substituted were those of the primary particles listed in Table 6.1. The data is representative of the entire range of Re_p^* and Ar values investigated to date, which spans particle diameters (d_p) of 5 nm to 3.5 mm at a particle density (ρ_p) of 3600 kg/m³ (alumina) or 5964 kg/m³ (zirconium). As mentioned earlier, the study on zirconium particles was included because it represented the highest values of Ar investigated to date. Fig. 6.6 indicates that, other than the well-acknowledged Zones I, II, and III, further Zones are plausible for the lower Re_p^* and Ar magnitudes, which thereby imposes a lower bound for Zone III. This is because of the varying inter-particle cohesion and hence agglomeration behaviors of the nano-scale particles due to the well-acknowledged effects of London-van der Waals, electrostatic, and moisture-induced surface tension forces (Hakim et al., 2005; Zhu et al., 2005). It should be noted that the pickup of agglomerates rather than individual particles has been acknowledged for Zone III (Kalman et al., 2005), but the nano-scale particles investigated in this study appear to suggest that the extents of agglomeration may be different. On the one hand, for the smallest particle diameters (d_p) investigated in this study of 5–135 nm, the agglomeration was so extensive such that the resulting agglomerates became large enough to behave like the micro-scale particles in Zone I. This proposed extrapolation of Zone I is labeled as Zone I' in Fig. 6.6. It should be noted that Zone I has been reported to be applicable for smaller Ar ranges for the pickup of particles in liquid-solid systems (Rabinovich and Kalman, 2007), but the underlying reasons are distinctly different. Whereas the extrapolation here was due to extensive cohesive effects leading to complex agglomerates, that in Rabinovich and Kalman (2007) was attributed to the negligible van der Waals forces in liquid-solid systems for the Ar range in which cohesive

forces are significant in gas-solid systems (Rabinovich and Kalman, 2007). On the other hand, for the intermediate particle diameters (d_p) investigated of 200–300 nm, a new Zone seems to exist in which the particles exhibited inter-particle cohesion effects intermediate between those of the original Zones III and I'. This new Zone is termed Zone IV, since it deviates from the available correlations defining the Zones. The intermediate behavior is presumably due to the coupled effects of increased van der Waals but decreased electrostatics interactions compared to that of Zone III, the same effects of which led to a weaker hierarchical structure of the agglomerates than that of Zone I'. Further studies on understanding the balance between the opposing effects of van der Waals and electrostatics interactions for nano-scale particles are necessary.

Fig. 6.6. Re_p^* vs Ar for particles (5 nm – 3,500,000 nm) representing the entire range of Re_p^* and Ar values investigated to date (Cabrejos and Klinzing, 1992; Hayden et al., 2003; Kalman et al., 2005) and Chapter 5. The solid lines represent correlations for Zones I, II, III and I' and the dotted line represents the newly proposed Zone IV. Each discrete datapoint represents each particle species investigated. Re_p^* (Eq. (2.5)) and Ar (Eq. (2.6)) were calculated using the individual particle diameter (d_p) listed in Table 6.1.

The correlation and bounds for Zones I and II remain unchanged. Under Zone I', a new range of particles at the lowest end of Re_p^* and Ar is suggested:

$$\text{Zone I': } Re_p^* = 5Ar^{\frac{3}{7}} \text{ for } Ar \leq 4 \times 10^{-7} \quad (6.2)$$

For Zone III, a new lower bound can be drawn, which can be expressed as:

$$\text{Zone III: } Re_p^* = 21.8Ar^{\frac{1}{3}} \text{ for } 5 \times 10^{-6} \leq Ar \leq 0.45 \quad (6.3)$$

As for the conceivable Zone IV in between Zones I' and III, the following correlation can be postulated:

$$\text{Zone IV: } \text{Re}_p^* = 1.3 \times 10^7 \text{Ar}^{\frac{10}{7}} \quad \text{for } 4 \times 10^{-7} < \text{Ar} < 5 \times 10^{-6} \quad (6.4)$$

Since only two of the particle samples investigated here fall into Zone IV, more data points would be beneficial to improving the correlation.

6.4 Conclusion

Experiments were conducted to determine the minimum pickup velocity (U_{pu}) of nine samples of alumina (Al_2O_3) particles with particle diameters (d_p) traversing the nano-scale and micro-scale (specifically in the range of 5–110,000 nm) by the weight loss method. This serves to reveal the missing link on the pneumatic conveying behavior of particles between $d_p = 21$ nm (Chapter 5) and $d_p = 530$ nm (Hayden et al., 2003) in terms of U_{pu} trends. The U_{pu} values obtained were then used to categorize the particles using the well-acknowledged Three-Zone classification (Kalman et al., 2005) .

The three key highlights are as follows. First, the minimum pickup velocity (U_{pu}) determined in this study exhibited a non-monotonic trend with respect to particle diameter (d_p) in the d_p range of 5 nm to 3.5 mm. Specifically, U_{pu} increased up to $d_p = 530$ nm, plateaued till $d_p = 15,000$ nm, then decreased till $d_p = 110,000$ nm before increasing again. Notably, the missing link revealed on the pneumatic conveying behavior of particles is that between $d_p = 5$ nm (i.e., nano-scale) and $d_p = 530$ nm (Hayden et al., 2003) (i.e., micro-scale) in terms of U_{pu} trends. Second, while the particles in the particle diameter (d_p) range of 5–135 nm agreed with the extrapolated Zone I correlation due to agglomeration effects, the two largest particle diameters (d_p) of 20,000 and 110,000 nm were expectedly classified, respectively, into Zones II and I; however, the intermediate particle diameters (d_p) of 200 and 300 nm could not be

classified into any of the three zones. Third, Zone III per se is inadequate in accounting for all the smaller cohesive particles, because of varying extents of inter-particle cohesion and hence agglomeration. For the particles investigated in this study which were expected to be categorized as Zone III, while the smallest ones (in this case, $d_p = 5\text{--}135\text{ nm}$) agglomerated so extensively such that the agglomerates became large enough to behave like the micro-scale particles in Zone I, the intermediate ones (in this case, $d_p = 200\text{--}300\text{ nm}$) did not agree with the Zone III correlation. Hence, within the original Zone III, a lower bound for Ar is suggested such that a new Zone IV can be labeled, along with Zone I' (i.e., the proposed extrapolation of Zone I) for the lowest ranges of Ar.

The findings of this study thus indicate that the minimum pickup velocity (U_{pu}) of nano-scale particles deviates from that expected from understanding based on micro-scale particles. This underscores the need for bridging the gap between the two scales.

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Part II

Fluidization

7

A comparative analysis of minimum fluidization velocity correlations

This chapter attempts to provide a comprehensive comparison of the U_{mf} values predicted by over a hundred correlations available in literature applied to Geldart Groups A, B and D.

The minimum fluidization velocity (U_{mf}), is defined as the superficial gas velocity at which the drag force of the upward moving gas is equal to the apparent weight of the particle bed (Fan and Zhu, 1998; Kunii and Levenspiel, 1991a). The experimental measurement is simpler and less subject to variations due to differences of reactors or instruments or analysis methods, since it is generally accepted as the superficial gas velocity at which the pressure drop across the particle bed plateaus or the expanded bed height starts increasing significantly. The standard procedure for experimentally determining U_{mf} is elucidated elsewhere (ASTM, 2012). The overall goal of the current study is to review the minimum fluidization velocity (U_{mf}) correlations available in literature in order to shed light on the state of such understandings and also provide perspectives for future research on this topic.

The U_{mf} value is important because it dictates the onset of fluidization (Fan and Zhu, 1998; Rhodes, 2008) and phenomena like the extent of segregation in bubbling fluidized beds (Chew and Hrenya, 2011; Chew et al., 2010; Cui and Grace, 2007; Rowe and Nienow, 1976; Tang and Puri, 2004). Over the past seven decades, ample efforts have been made to improve the understanding of U_{mf} , as shown in Fig. 7.1, which reflects the number of publications related to U_{mf} and the number of correlations developed per decade since 1950. More than a hundred

empirical and semi-empirical correlations for determining U_{mf} are available in literature. Generally, U_{mf} is expressed as a function of particle properties (e.g., particle diameter (d_p), particle density (ρ_p), particle sphericity (ϕ)), gas properties (e.g., gas density (ρ), gas viscosity (μ)) and bed properties (e.g., bed voidage at minimum fluidization (ε_{mf})). Three important observations regarding Fig. 7.1 are as follows: (i) the number of publications related to U_{mf} have sharply risen in the last two decades, indicating a recent renewed interest, presumably due to the growing popularity of biomass fluidization; (ii) the number of simulation studies are more than experimental studies in the current decade and have almost tripled since the last decade, reflecting the growing computing power, as also seen in the study of particle clusters (Cahyadi et al., 2017); and (iii) the number of new U_{mf} correlations per decade have increased till the 1980 decade and remained relatively constant ever since. Regarding (iii), the percentage of new U_{mf} correlations published relative to the total number of publications per decade is reducing, which suggests a possible awareness in the limited applicability of such correlations, and that researchers are turning towards a more physical-based understanding of U_{mf} increasingly through simulations or comparison of results to existing correlations.

Fig. 7.1. Number of experimental and simulation studies and new correlations for minimum fluidization velocity (U_{mf}) per decade from 1950. Source = Scopus. Keyword search = ((minimum AND fluidization) OR (minimum AND fluidization) OR (incipient AND fluidization) OR (incipient AND fluidization)) AND (velocity) AND NOT (reactor OR kinetic). Updated on 13 August 2017.

In sync with previous reviews (Cahyadi et al., 2015; Chew et al., 2015), the predicted U_{mf} values compared within each Geldart Group, then plausible reasons underlying the discrepancies are described. This study thus endeavors to advance the understanding or lack thereof of U_{mf} through a comparative analysis of the correlations available in literature. To better delineate the scope of this study, it should be noted that this review does not: (i) report new experimental data to validate available correlations, and hence cannot comment on which correlations are right or wrong, or succeed or fail in predicting U_{mf} accurately; (ii) make

recommendations on which correlation is the ‘best’, since such clarity requires more physical understanding and description of the incipient fluidization phenomenon, which is beyond the current scope and (iii) present any new correlations, since this would be an antithesis to the essence of this work given the current physical understanding on U_{mf} . Instead, the review seeks to inform on the discrepancies associated with the predictions of U_{mf} values, and attempt to highlight common pitfalls and recommend future studies towards improving such predictions. Due to the large number of correlations and the similarity in the expressions, the correlations are categorized into four groups according to the form of the correlation.

More than a hundred correlations for predicting U_{mf} are available in the literature. Most of them are functions of the dimensionless Reynolds (Re_{mf}) and Archimedes (Ar) numbers. Re_{mf} , the Reynolds number at minimum fluidization is expressed as:

$$Re_{mf} = \frac{\rho_p d_p U_{mf}}{\mu} \quad (7.1)$$

Ar is explained in Chapter 2; the expression is reiterated here for convenience.

$$Ar = \frac{g \rho (\rho_p - \rho) d_p^3}{\mu^2} \quad (2.6)$$

where ρ_p is the particle density, d_p is the particle diameter, μ is the ambient air viscosity, ρ is the density of ambient air and g is the gravitational acceleration.

The general forms of the correlations can be categorized into four key embodiments:

1. $Re_{mf} = (K_1^2 + K_2 Ar)^{0.5} - K_1$, where K_1 and K_2 are empirical constants
2. $Re_{mf} = A \cdot Ar^b$, where A and b are empirical constants
3. $Re_{mf} = \frac{Ar}{p + q\sqrt{Ar}}$, where p and q are empirical constants

4. $U_{mf} = K X^\alpha$, where $X = \left(\frac{d_p^2(\rho_p - \rho)g}{\mu} \left(\frac{\rho_p}{\rho} \right)^{1.23} \right)$, and K and α are empirical constants

Accordingly, the ensuing four sections are dedicated to the four categories. Within each section, the correlations are applied to Geldart (1973) groups A, B and D to assess the span of the predicted U_{mf} values. It was noted in a previous review on entrainment correlations (Chew et al., 2015) that whether a particular correlation was developed for a specific Geldart Group or not seems inconsequential, since outlying entrainment predictions do not necessarily imply application to a different Geldart Group than was developed for; therefore, the current study also attempted to apply all correlations to each Geldart Group. Continuing this line of reasoning, the particle properties considered for the base case parameters (Tables 7.2, 7.3 and 7.4) of the various Geldart Groups for evaluating the U_{mf} correlations were those of the more industrially relevant or popular particles in that Geldart Group (Fluid Catalytic Cracking, FCC catalysts for Geldart Group A, sand for Geldart Group B and coal for Geldart Group D). Particularly regarding the parameters of particle sphericity (ϕ) and voidage at minimum fluidization (ε_{mf}), whose values are not readily available, most authors have (i) not reported their values, (ii) reported them only qualitatively, e.g. spherical or non-spherical particles, and/or (iii) included their effects in the U_{mf} correlations in the form of empirical coefficients. However, for those correlations which have included them, a fifth section is added to evaluate the sensitivity of the correlations to these difficult-to-measure parameters of ϕ and ε_{mf} . The gas phase is always air, with a density (ρ) and viscosity (μ) of 1.2 kg/m³ and 0.000018 kg/s/s, respectively.

7.1 $\text{Re}_{mf} = (K_1^2 + K_2 \text{Ar})^{0.5} - K_1$ (Wen and Yu (1966)) type of correlations

One of the earliest correlations developed to determine minimum fluidization velocity (U_{mf}) was based on that to predict the pressure drop across a packed bed by Ergun (1952),

which was in turn based on the Kozeny-Carman equation (Carman, 1937; Kozeny, 1927). The Ergun equation, which has been popularly used for calculating the minimum fluidization velocity (U_{mf}), is a semi-empirical correlation accounting for the effects of particle diameter (d_p), particle density (ρ_p), gas density (ρ), gas viscosity (μ), particle sphericity (ϕ) and bed voidage at minimum fluidization (ε_{mf}) (Kunii and Levenspiel, 1991b) :

$$\frac{1.75}{\varepsilon_{mf}^3 \phi} \text{Re}_{mf}^2 + \frac{150(1-\varepsilon_{mf})}{\varepsilon_{mf}^3 \phi^2} \text{Re}_{mf} = \text{Ar} \quad (7.2)$$

Since the values of ϕ and ε_{mf} are typically difficult to determine with precision, Eq. (7.2) can be re-expressed as follows:

$$C_1 \text{Re}_{mf}^2 + C_2 \text{Re}_{mf} = \text{Ar} \quad (7.3)$$

where

$$C_1 = \frac{1.75}{\varepsilon_{mf}^3 \phi} \quad (7.4)$$

$$C_2 = \frac{150(1-\varepsilon_{mf})}{\varepsilon_{mf}^3 \phi^2} \quad (7.5)$$

Wen and Yu (1966) represented one of the earliest studies on modifying the Ergun equation per Eq. (7.3) to calculate U_{mf} . They found that K_1 and K_2 were invariant over a wide range of Re_{mf} of 0.001 – 4000, thus Eq. (7.3) was expressed as:

$$\text{Re}_{mf} = (33.7^2 + 0.0408\text{Ar})^{0.5} - 33.7 \quad (7.6)$$

where $C_2/2C_1 = 33.7$ and $1/C_1 = 0.0408$. This form was adopted by numerous researchers subsequently:

$$\text{Re}_{mf} = (K_1^2 + K_2\text{Ar})^{0.5} - K_1 \quad (7.7)$$

Table 7.1 presents all the correlations that took the form of Eq. (7.7) for predicting U_{mf} chronologically from 1952 to 2016. The ranges of particle diameters (d_p), particle densities (ρ_p) and Geldart Groups investigated for each correlation are also listed in Table 7.1 where available.

Table 7.1. $Re_{mf} = (K_1^2 + K_2 Ar)^{0.5} - K_1$ (Wen and Yu (1966)) type of correlations. The ranges of particle diameters (d_p), particle densities (ρ_p) and Geldart Groups investigated for each correlation are also listed where available.

Author(s)	Correlation	Particle diameter, d_p (μm)	Particle density, ρ_p (kg/m^3)	Geldart Group(s)
Ergun (1952)	$Re_{mf} = \left(\left(\frac{42.86(1 - \varepsilon_{mf})}{\phi} \right)^2 + 0.571\varepsilon_{mf}^3 \phi Ar \right)^{0.5} - \left(\frac{42.86(1 - \varepsilon_{mf})}{\phi} \right)$	250-1190	1060-1270	B, D
Wen and Yu (1966)	$Re_{mf} = (33.7^2 + 0.0408Ar)^{0.5} - 33.7$	2052-6350	2360-7840	D
Bourgeois and Grenier (1968)	$Re_{mf} = (25.46^2 + 0.0382Ar)^{0.5} - 25.46$	86-25000	1200-19300	A, B, D
Saxena and Vogel (1977)	$Re_{mf} = (25.28^2 + 0.0571Ar)^{0.5} - 25.28$	650-704	1900-2460	B, D
Babu et al. (1978)	$Re_{mf} = (25.25^2 + 0.0651Ar)^{0.5} - 25.25$	50-2870	2560-3920	A, B, D
Vaid and Sen Gupta (1978)	$Re_{mf} = (24^2 + 0.0546Ar)^{0.5} - 24$	114-1829	1669-4332	B, D
Richardson and da S. Jerónimo (1979)	$Re_{mf} = (25.7^2 + 0.0365Ar)^{0.5} - 25.7$			

Author(s)	Correlation	Particle diameter, d_p (μm)	Particle density, ρ_p (kg/m^3)	Geldart Group(s)
Pattipati and Wen (1981)	$\text{Re}_{\text{mf}} = \left([83.33(1 - \epsilon_{\text{mf}})]^2 + 0.417\epsilon_{\text{mf}}^3 \text{Ar} \right)^{0.5} - [83.33(1 - \epsilon_{\text{mf}})]$	240-3376	2630	B, D
Grace (1982)	$\text{Re}_{\text{mf}} = (27.2^2 + 0.0408\text{Ar})^{0.5} - 27.2$			
Chitester et al. (1984)	$\text{Re}_{\text{mf}} = (28.7^2 + 0.0494\text{Ar})^{0.5} - 28.7$	88-374	1.12-2.47	A, B
Thonglimp et al. (1984a)	$\text{Re}_{\text{mf}} = (31.6^2 + 0.0425\text{Ar})^{0.5} - 31.6$	112-2125	1.6-7.4	B, D
Thonglimp et al. (1984b)	$\text{Re}_{\text{mf}} = (19.9^2 + 0.03196\text{Ar})^{0.5} - 33.7$			
Zheng et al. (1985)	$\text{Re}_{\text{mf}} = (18.75^2 + 0.03125\text{Ar})^{0.5} - 18.75$	350-1410	2.52-2.65	B, D
Nakamura et al. (1985)	$\text{Re}_{\text{mf}} = (33.95^2 + 0.0465\text{Ar})^{0.5} - 33.95$	200-4000		B, D
Noda et al. (1986) 1	$\text{Re}_{\text{mf}} = (19.29^2 + 0.0276\text{Ar})^{0.5} - 19.29$ Mixed bed post-fluidization	454-12500	440-7530	B, D
Noda et al. (1986) 2	$\text{Re}_{\text{mf}} = (88.99^2 + 0.0276\text{Ar})^{0.5} - 88.99$ Segregated bed post-fluidization, $d_{p, \text{big}}/d_{p, \text{small}} > 3$			
Lucas et al. (1986) 1	$\text{Re}_{\text{mf}} = (29.5^2 + 0.0357\text{Ar})^{0.5} - 29.5,$ $0.8 < \varphi < 1$			
Lucas et al. (1986) 2	$\text{Re}_{\text{mf}} = (32.1^2 + 0.0571\text{Ar})^{0.5} - 32.1,$ $0.5 < \varphi < 0.8$			

Author(s)	Correlation	Particle diameter, d_p (μm)	Particle density, ρ_p (kg/m^3)	Geldart Group(s)
Lucas et al. (1986) 3	$\text{Re}_{\text{mf}} = (25.2^2 + 0.0672\text{Ar})^{0.5} - 25.2,$ $\varphi < 0.5$			
Chyang and Huang (1988)	$\text{Re}_{\text{mf}} = (33.3^2 + 0.033\text{Ar})^{0.5} - 33.3$	699-6062	910-6860	B, D
Sathyanarayana and Rao (1989)	$\text{Re}_{\text{mf}} = (30.1^2 + 0.0417\text{Ar})^{0.5} - 30.1$	450-1700		B, D
Panigrahi and Murty (1991)	$\text{Re}_{\text{mf}} = (32.2^2 + 0.0382\text{Ar})^{0.5} - 32.2$			
Wu and Baeyens (1991)	$\text{Re}_{\text{mf}} = (30.85^2 + 0.0379\text{Ar})^{0.5} - 30.85$	104-1970	1.6-2.6	B, D
Adánez and Abanades (1991) 1	$\text{Re}_{\text{mf}} = (25.18^2 + 0.0373\text{Ar})^{0.5} - 25.18$	502-2828	1.48-2.60	B, D
Adánez and Abanades (1991) 2	$\text{Re}_{\text{mf}} = (9.88^2 + 0.0297\text{Ar})^{0.5} - 9.88$	710-3578	1.00-1.61	B, D
Tannous (1993)	$\text{Re}_{\text{mf}} = (23.785^2 + 0.0413\text{Ar})^{0.5} - 23.785$			
Biń (1994)	$\text{Re}_{\text{mf}} = (27.31^2 + 0.0386\text{Ar})^{0.5} - 27.31$	40-2120	1600-7500	A, B, D
Tannous et al. (1994) 1	$\text{Re}_{\text{mf}} = \left[\left(33.3 \frac{1 - \varepsilon_{\text{mf}}}{\varphi} \right)^2 + 0.575 \varphi \varepsilon_{\text{mf}}^3 \text{Ar} \right]^{0.5} - \left(33.3 \frac{1 - \varepsilon_{\text{mf}}}{\varphi} \right)$	715-7500	1016-3950	B, D
Tannous et al. (1994) 2	$\text{Re}_{\text{mf}} = (25.83^2 + 0.043\text{Ar})^{0.5} - 25.83$			

Author(s)	Correlation	Particle diameter, d_p (μm)	Particle density, ρ_p (kg/m^3)	Geldart Group(s)
Llop et al. (1996) 1	$\text{Re}_{\text{mf}} = (29.4^2 + 0.0357\text{Ar})^{0.5} - 29.4$ $0.8 < \varphi < 1$ (no slip condition)	213-1600	1600-2650	B, D
Llop et al. (1996) 2	$\text{Re}_{\text{mf}} = (38.6^2 + 0.0571\text{Ar})^{0.5} - 38.6$ $0.5 < \varphi < 0.8$ (no slip condition)			
Reina et al. (2000)	$\text{Re}_{\text{mf}} = (48^2 + 0.045\text{Ar})^{0.5} - 48$	1140-1690	500-620	B, D
Hartman et al. (2000) 1	$\text{Re}_{\text{mf}} = (17.32^2 + 0.0216\text{Ar})^{0.5} - 17.32$	125-800	2900	B, D
Hartman et al. (2000) 2	$\text{Re}_{\text{mf}} = (7.03^2 + 0.0101\text{Ar})^{0.5} - 7.03$	125-800	1700	A, B, D
Hilal et al. (2001)	$\text{Re}_{\text{mf}} = (13.07^2 + 0.0263\text{Ar})^{0.5} - 13.07$	80-1230	1228-8900	A, B, D
Kozanoglu et al. (2002)	$\text{Re}_{\text{mf}} = (33.6^2 + 0.0598\text{Ar})^{0.5} - 33.6$	2250-7730	668-1414	D
Delebarre (2004)	$\text{Re}_{\text{mf}} = \left((600\varepsilon_{mf}^3(1 - \varepsilon_{mf}))^2 + 0.0408\text{Ar} \right)^{0.5} - (600\varepsilon_{mf}^3(1 - \varepsilon_{mf}))$			
Zhiping et al. (2007)	$\text{Re}_{\text{mf}} = (22.1^2 + 0.0354\text{Ar})^{0.5} - 22.1$	300-2000	2.497-2.582	B, D
Hartman et al. (2007)	$\text{Re}_{\text{mf}} = (21.32^2 + 0.02833\text{Ar})^{0.5} - 21.32$	650-5300	902.7-1149	B, D
Mohanta et al. (2012)	$\text{Re}_{\text{mf}} = (41.96^2 + 0.049\text{Ar})^{0.5} - 41.96$	33-1070	4700	A, B, D

Author(s)	Correlation	Particle diameter, d_p (μm)	Particle density, ρ_p (kg/m^3)	Geldart Group(s)
Paudel and Feng (2013) 1	$\text{Re}_{\text{mf}} = (30.28^2 + 0.0464\text{Ar})^{0.5} - 30.28$	241-490	2500-3940	B
Paudel and Feng (2013) 2	$\text{Re}_{\text{mf}} = (30.28^2 + 0.108\text{Ar})^{0.5} - 30.28$	856-1040	1080-1200	B, D
Kumoro et al. (2014)	$\text{Re}_{\text{mf}} = ((30.81\varphi^2)^2 + 0.0674\text{Ar})^{0.5} - 30.81\varphi^2$	241-1560	635-2630	B, D
Tannous and Lourenço (2015)	$\text{Re}_{\text{mf}} = (33.9^2 + 0.051\text{Ar})^{0.5} - 33.9$	331-2017	395-2636	A, B, D
Zarekar et al. (2016)	$\text{Re}_{\text{mf}} = \left(\left(\frac{25.7}{\varphi} \right)^2 + 0.0365\varphi\text{Ar} \right)^{0.5} - \frac{25.7}{\varphi}$ (no slip boundary condition at wall)	120-728	2500	B, D

7.1.1 $\text{Re}_{\text{mf}} = (K_1^2 + K_2\text{Ar})^{0.5} - K_1$ type of correlations: Geldart Group A

This section evaluates all the correlations listed in Table 7.1 for the most industrially relevant Geldart Group A particles, namely, the fluid catalytic cracking (FCC) catalysts. Specifically, the U_{mf} values predicted by all such correlations are compared across a range of particle diameters (d_p) while keeping all other parameters constant. Table 7.2 gives the base case parameters assessed, the particle properties of which mimic the Fluid Catalytic Cracking (FCC) catalyst particles.

Table 7.2. Base case parameters for Geldart Group A particles

Parameter	Symbol	Value	Unit
Diameter	d_p	30-170	μm
Density	ρ_p	1450	kg/m^3
Sphericity	ϕ	0.58	-
Voidage at minimum fluidization	ε_{mf}	0.42	-

Fig. 7.2 displays a semi-log plot of U_{mf} calculated using the correlations in Table 7.1 against particle diameter (d_p) for Geldart group A particles. Because all of over 40 correlations hampers the readability of the graph, in all subsequent figures, correlations that superimpose on one another have been replaced by representative ones.

Fig. 7.2. $\text{Re}_{mf} = (K_1^2 + K_2 \text{Ar})^{0.5} - K_1$ type of correlations: Semi-log plot of U_{mf} vs. d_p for Geldart Group A particles

Fig. 7.2 indicates that the discrepancy among the U_{mf} predictions by the correlations in this category (Table 7.1) is one order-of-magnitude, which warrants a more detailed assessment

of the underlying reasons. Firstly, the shapes of the curves are all similar, with U_{mf} increasing rapidly initially and then more gradually with d_p . The increase of U_{mf} values by one order-of-magnitude in the d_p range of 30 and 170 μm investigated is because the dimensionless number Ar is proportional to the cube of d_p (i.e., d_p^3), while the decreasing gradients are due to the subtraction of the empirical constant K_I in such correlations. Secondly, the lowest U_{mf} values are given by Noda et al. (1986), which is presumably because the U_{mf} measured for the mixture of Geldart Group B and low-density (400-1200 kg/m^3) Geldart Group D particles considered was that at incipient fluidization rather than complete fluidization. The same authors (Noda et al., 1986) have further developed a correlation based on the complete fluidization velocity, which agrees more with the other correlations. The Ergun (1952) correlation gives the second lowest prediction, presumably since it has been developed for Geldart Groups B and D particles and the inter-particle cohesion of Group A particles, which would increase U_{mf} , is not considered. Another correlation that also gives one of the lowest predictions of U_{mf} is that by Pattipati and Wen (1981), which is likely because of the higher temperatures investigated (namely, 25 - 850 $^{\circ}\text{C}$). Fourthly, the correlations by Kumoro et al. (2014), Paudel and Feng (2013), Adánez and Abanades (1991), and Saxena and Vogel (1977) give the higher U_{mf} predictions, likely because these investigations were on biomass or fuel particles, which are classified as low-density Geldart Group B and Geldart Group D particles. In particular, the Kumoro et al. (2014) correlation predicts the highest U_{mf} values due to the term ϕ^2 in K_I , which significantly reduces the value of K_I and thereby increases the value of U_{mf} . Among other correlations predicting higher U_{mf} values, the Babu et al. (1978) correlation over-predicts U_{mf} despite having considered Geldart Group A, B and D particles, but the underlying reason cannot be established since the details of the particles are not available; and the Lucas et al. (1986) correlation over-predicts likely due to the highly non-spherical particles ($\phi < 0.5$) investigated.

7.1.2 $Re_{mf} = (K_1^2 + K_2 Ar)^{0.5} - K_1$ type of correlations: Geldart Group B

This section evaluates all available $Re_{mf} = (K_1^2 + K_2 Ar)^{0.5} - K_1$ type of correlations (Table 7.1) for Geldart Group B particles, the properties of which are listed in Table 7.3. The particle properties are those of sand, which is a common representation of Geldart Group B particles.

Table 7.3. Base case parameters for Geldart Group B particles

Parameter	Symbol	Value	Unit
Diameter	d_p	90-650	μm
Density	ρ_p	2650	kg/m^3
Sphericity	ϕ	0.85	-
Voidage at minimum fluidization	ε_{mf}	0.42	-

Fig. 7.3. $\text{Re}_{mf} = (K_1^2 + K_2 \text{Ar})^{0.5} - K_1$ type of correlations: Semi-log plot of U_{mf} vs. d_p for Geldart Group B particles

Fig. 7.3 displays the predicted U_{mf} values with respect to particle diameter (d_p) for Geldart Group B particles. The trends are similar to that for Geldart Group A particles in Fig. 7.2 particularly on two counts. Firstly, all correlations predict a monotonic increase in U_{mf} with

an increase in d_p , and the disparity of U_{mf} values for the range of d_p values investigated is about an order-of-magnitude. Secondly, Noda et al. (1986) and Pattipati and Wen (1981) persisted in giving lower U_{mf} predictions, presumably due to similar reasons. Paudel and Feng (2013) and Kumoro et al. (2014) persisted in giving higher U_{mf} predictions, but the latter gave lower U_{mf} values than the former due to the influence of the ϕ^2 term in K_I of the latter decreasing with increasing ϕ . (Ergun, 1952) which gives lower U_{mf} predictions for Group A gives reasonable predictions for the non-cohesive Group B particles, while (Reina et al., 2000) predicts lower U_{mf} values for Group B likely because it was developed from low- ρ_p (500-620 kg/m³) Group B and Group D data. The (Saxena and Vogel, 1977) and (Lucas et al., 1986) correlations give similarly high U_{mf} predictions for Geldart Group B likely due to similar reasons of high d_p and low ϕ , respectively. The (Babu et al., 1978) correlation also maintains a similar discrepancy of predicted U_{mf} values relative to other correlations as Group A.

7.1.3 $Re_{mf} = (K_1^2 + K_2 Ar)^{0.5} - K_1$ type of correlations: Geldart Group D

This section evaluates all available Wen and Yu type of correlations for Geldart Group D, which is commonly represented by coal, whose particle properties are given in Table 7.4.

Table 7.4. Base case parameters for Geldart Group D particles

Parameter	Symbol	Value	Unit
Diameter	d_p	800-3200	μm
Density	ρ_p	1550	kg/m ³
Sphericity	ϕ	0.70	-
Voidage at minimum fluidization	ε_{mf}	0.45	-

Fig. 7.4. $\text{Re}_{mf} = (K_1^2 + K_2 \text{Ar})^{0.5} - K_1$ type of correlations: Semi-log plot of U_{mf} vs. d_p for Geldart Group D

Fig. 7.4 displays the U_{mf} versus d_p plots when the correlations are applied to Geldart group D particles. Two similarities are clear among the trends for all three Geldart Groups (Figs. 7.2-7.4). One is that the highest U_{mf} values are by (Paudel and Feng, 2013) and (Kumoro et al., 2014). The over-prediction has been attributed to the investigation of Geldart Group D

particles earlier, so it is surprising that the over-prediction persists even when applied to Geldart Group D particles in Fig 7.2. It could thus be that the limited studies on Geldart Group D particles precludes any generalization. While (Paudel and Feng, 2013) investigated low-density, high-voidage biomass particles (namely, corn cob and walnut shell), Kumoro et al. (2014) used highly irregularly-shaped ($\phi = 0.18-0.71$) biomass particles (rice husk and corn cob). Another is that the Noda et al. (1986) correlation persists in giving very low U_{mf} values, which is due to the reporting of incipient rather than complete fluidization velocity for the binary-size mixture investigated. In addition, three interesting contrasts between Fig. 7.4 for the Geldart Group D particles, and Figs. 7.2 and 7.3 for Geldart Groups A and B, respectively, are observed. Firstly, the discrepancy among the predicted U_{mf} values is clearly lesser for Geldart Group D (Fig. 7.4), compared to that for Geldart Groups A (Fig. 7.2) and B (Fig. 7.3). While the greatest difference between the highest and lowest predictions for Group D is fourfold, that for Geldart Groups A and B is an order-of-magnitude. Secondly, regarding the shapes of the trends, the trends for Geldart Group D are much flatter than those of Geldart Groups A and B. This can be attributed to the decreasing influence of the subtraction of K_I from $(K_I^2 + K_2 Ar)^{0.5}$ in the expression (Eq. (7.7)). Thirdly, the Hartman et al. (2000) correlation, which does not give outlying U_{mf} values when applied to Geldart Groups A and B, gives significantly low U_{mf} values for Geldart Group D particles. This is presumably due to the high value for the constant K_I (Eq. (7.7)), resulting from a dataset having a low d_p (125-800 μm) compared to the test case ($d_p = 800-3200 \mu\text{m}$) Geldart Group D particles.

7.1.4 Summary

Applying the $\text{Re}_{mf} = (K_1^2 + K_2 Ar)^{0.5} - K_1$ type of correlations to representative particles of Geldart Groups A, B and D reveals three key highlights: (i) the U_{mf} versus d_p trends are similar for all three Groups; (ii) the difference between the maximum and minimum U_{mf} predictions is an order-of-magnitude for all three Groups; (iii) the largest discrepancy among

the predictions is for Geldart Group A particles presumably due to a lack of understanding of inter-particle cohesive effects, while the least is for Geldart Group D particles presumably due to the specific form of this type of correlations (Eq. (7.7)).

7.2 $\text{Re}_{\text{mf}} = A \cdot \text{Ar}^b$ type of correlations

This type of correlations is the second most abundant in literature. More than 40 correlations of this type are available, as given chronologically in Table 7.5. They have the general format:

$$\text{Re}_{\text{mf}} = A \cdot f(\varphi, \varepsilon_{\text{mf}}) \cdot [g(\rho_p, \rho)]^x \cdot \text{Ar}^b \quad (7.8)$$

where Re_{mf} is the Reynolds number at minimum fluidization, ε_{mf} is the bed voidage at minimum fluidization, φ is the particle sphericity, Ar is the Archimedes number and A , x and b are empirical constants. To eliminate the need to experimentally determine bed voidage (ε_{mf}) and particle sphericity (φ) and simplify the expression, ε_{mf} and φ are typically incorporated into empirical constants A and b via data-fitting to obtain the best regression coefficients, and assume the following format:

$$\text{Re}_{\text{mf}} = A \cdot \text{Ar}^b \quad (7.9)$$

Due to the regression of the experimental data, such correlations are inevitably somewhat system-specific and not readily applicable to beyond the scope investigated. Many of these correlations designate ‘ b ’ with a value of unity.

Table 7.5. $Re_{mf} = A \cdot Ar^b$ correlations. The ranges of particle diameters (d_p), particle densities (ρ_p) and Geldart Groups investigated for each correlation are also listed where available.

Reference Author(s)	Correlation	Particle diameter, d_p (μm)	Particle density, ρ_p (kg/m^3)	Geldart Group(s)
Johnson (1949-50)	$Re_{mf} = 0.056 \left(\frac{\varphi^2 \varepsilon_{mf}^5}{1+0.5(1-\varepsilon_{mf})} \right) Ar,$ $Re < 2$			
Miller and Logwinuk (1951)	$Re_{mf} = 0.00125 \left(\frac{\rho}{\rho_p - \rho} \right)^{0.1} Ar$	97-249	1120-3890	A, B
Leva (1959)	$Re_{mf} = 0.000822 Ar^{0.94}$	51-970	1600-2700	A, B, D
Rowe and Henwood (1961)	$Re_{mf} = 0.0081 Ar$	12700-15875	~1000	D
Frantz (1966)	$Re_{mf} = 0.001065 Ar$	46-305		A, B
Davies and Richardson (1966)	$Re_{mf} = 0.00078 Ar$	55-142	945-1271	A
Pillai and Rao (1971)	$Re_{mf} = 0.000701 Ar$	58-1100		A, B, D
Baeyens and Geldart (1973)	$Re_{mf} = 0.0009125 Ar^{0.934}$	50-4000	850-8810	A, B, D
Kumar and Sen Gupta (1974)	$Re_{mf} = 0.0054 Ar^{0.78}$	359-677	2660-4570	B, D
Riba et al. (1978)	$Re_{mf} = 0.0154 \left(\frac{\rho}{\rho_p - \rho} \right)^{-0.04} Ar^{0.66}$	600-10360	1342-7741	B, D
Doichev and Akhnikov (1979)	$Re_{mf} = 0.00097 Ar^{0.947}$	90-2200	2650	B, D

Reference Author(s)	Correlation	Particle diameter, d_p (μm)	Particle density, ρ_p (kg/m^3)	Geldart Group(s)
Maeno and Nishimura (1979)	$\text{Re}_{\text{mf}} = \frac{\varepsilon_{\text{mf}}^3}{150(1 - \varepsilon_{\text{mf}})} \text{Ar}$	300-1950		B, D
Thonglimp et al. (1984a) 1	$\text{Re}_{\text{mf}} = 0.000754 \text{Ar}^{0.98},$ $\text{Re}_{\text{mf}} < 30$	112-2125	1600- 7400	B, D
Thonglimp et al. (1984a) 2	$\text{Re}_{\text{mf}} = 0.0195 \text{Ar}^{0.66},$ $\text{Re}_{\text{mf}} > 30$			
Thonglimp et al. (1984b) 1	$\text{Re}_{\text{mf}} = 0.0034 \left(\frac{\rho}{\rho_p - \rho} \right)^{0.1} \text{Ar}^{0.9},$ $\text{Re}_{\text{mf}} < 20$			
Thonglimp et al. (1984b) 2	$\text{Re}_{\text{mf}} = 0.0279 \text{Ar}^{0.63}$ $\text{Re}_{\text{mf}} > 20$			
Fatah (1991)	$\text{Re}_{\text{mf}} = 0.0459 \text{Ar}^{0.6116}$	670-3570	3950	D
Fletcher et al. (1992)	$\text{Re}_{\text{mf}} = 0.000714 \text{Ar},$ $\varphi > 0.9, \text{Ar} < 1480$	75-2320	~2500	A, B, D
Mourad et al. (1994) 1	$\text{Re}_{\text{mf}} = 0.000552 \text{Ar}^{1.044}$ Apparent fluidization	250-8000	1200- 2650	B, D
Mourad et al. (1994) 2	$\text{Re}_{\text{mf}} = 0.00106 \text{Ar}^{1.04}$ Complete fluidization			
Xie and Geldart (1995)	$\text{Re}_{\text{mf}} = \frac{\varepsilon_{\text{mf}}^3}{180(1 - \varepsilon_{\text{mf}})} \text{Ar}$	26-137	1210- 1420	A
Barbosa et al. (1995)	$\text{Re}_{\text{mf}} = 0.0019 \text{Ar}^{0.87}$	110-920	1480- 3960	B, D
Gauthier et al. (1999) 1	$\text{Re}_{\text{mf}} = 0.0022 \text{Ar}^{0.818}$ $0.78 < \text{Re}_{\text{mf}} < 78 \text{ and}$ $1800 < \text{Ar} < 500000$	160-2500		B, D

Reference Author(s)	Correlation	Particle diameter, d_p (μm)	Particle density, ρ_p (kg/m^3)	Geldart Group(s)
Gauthier et al. (1999) 2	$\text{Re}_{\text{mf}} = 0.0052\text{Ar}^{0.777}$ $1.35 < \text{Re}_{\text{mf}} < 113$ and $1800 < \text{Ar} < 500000$			
Subramani et al. (2007) 1	$\text{Re}_{\text{mf}} = 0.000606\text{Ar}$ $(1 - \varepsilon_{\text{mf}})^{-1} \text{Re}_{\text{mf}} < 10,$ $\varepsilon_{\text{mf}} < 0.5,$ Room temperature	128-200	2636-4690	B
Subramani et al. (2007) 2	$\text{Re}_{\text{mf}} = 0.000666\text{Ar}$ Temperature: 250-900 K			
Subramani et al. (2007) 3	$\text{Re}_{\text{mf}} = \left(\frac{0.3507}{\varepsilon_{\text{mf}}}\right)^{14.2} \text{Ar}^{0.55}$ Temperature: 250-900 K			
Rabinovich and Kalman (2008) D	$\text{Re}_{\text{mf}} = 0.015\text{Ar}^{0.667}$ For Geldart Group D			D
Rabinovich and Kalman (2008) B	$\text{Re}_{\text{mf}} = 0.0013\text{Ar}^{0.88}$ For Geldart Group B			B
Goo et al. (2009)	$\text{Re}_{\text{mf}} = 0.0005999\text{Ar}$	210-380	2500	B
Chen et al. (2010)	$\text{Re}_{\text{mf}} = 0.01036\text{Ar}^{0.7107}$	180-450		B
de Vasconcelos and Mesquita (2011)	Re_{mf} $= 0.2029 \left(\frac{\varphi}{\varepsilon_{\text{mf}}}\right)^{0.7} \left(\frac{\rho}{\rho_p - \rho}\right)^{0.5}$ $\text{Ar}^{0.75}$	18-405	3263-3387	A, B

Reference Author(s)	Correlation	Particle diameter, d_p (μm)	Particle density, ρ_p (kg/m^3)	Geldart Group(s)
Jiliang et al. (2013)	Re_{mf} $= 0.0829 \left(\frac{\rho_p}{\rho_p - \rho} \right)^{0.533} \text{Ar}^{0.533}$	500-1290	2500- 2750	B, D
Puspasari et al. (2013) A1	Re_{mf} $= 3.92$ $\cdot 10^{-6} \varphi^{1.826} (1$ $- \varepsilon_{\text{mf}})^{0.942} \left(\frac{\rho_p}{\rho} \right)^{0.942} \text{Ar}^{0.942}$ $6 \leq \ln \left[\varphi^3 (1 - \varepsilon_{\text{mf}}) \left(\frac{\rho_p}{\rho} \right) \text{Ar} \right]$ ≤ 9 <p>For Geldart Group A</p>	33-124	790-907	A
Puspasari et al. (2013) B1	Re_{mf} $= 7.66$ $\cdot 10^{-10} \varphi^{3.734} (1$ $- \varepsilon_{\text{mf}})^{1.578} \left(\frac{\rho_p}{\rho} \right)^{1.578} \text{Ar}^{1.578}$ $12.5 \leq \ln \left[\varphi^3 (1 - \varepsilon_{\text{mf}}) \left(\frac{\rho_p}{\rho} \right) \text{Ar} \right]$ ≤ 15 <p>For Geldart Group B</p>	167-715	1330- 3220	B

Reference Author(s)	Correlation	Particle diameter, d_p (μm)	Particle density, ρ_p (kg/m ³)	Geldart Group(s)
Puspasari et al. (2013) B2	$\text{Re}_{mf} = 7.02 \cdot 10^{-7} \varphi^{1.958} (1 - \varepsilon_{mf})^{0.986} \left(\frac{\rho_p}{\rho}\right)^{0.986} \text{Ar}^{0.986}$ $15 < \ln \left[\varphi^3 (1 - \varepsilon_{mf}) \left(\frac{\rho_p}{\rho}\right) \text{Ar} \right] \leq 16.5$ <p>For Geldart Group B</p>	167-715	1330-3220	B
Puspasari et al. (2013) D1	$\text{Re}_{mf} = 2.42 \varphi^{-0.415} (1 - \varepsilon_{mf})^{0.195} \left(\frac{\rho_p}{\rho}\right)^{0.195} \text{Ar}^{0.195}$ $15 \leq \ln \left[\varphi^3 (1 - \varepsilon_{mf}) \left(\frac{\rho_p}{\rho}\right) \text{Ar} \right] \leq 17$ <p>For Geldart Group D</p>	745-6911	619-2616	D
Puspasari et al. (2013) D2	$\text{Re}_{mf} = 6.96 \cdot 10^{-4} \varphi^{0.77} (1 - \varepsilon_{mf})^{0.59} \left(\frac{\rho_p}{\rho}\right)^{0.59} \text{Ar}^{0.59}$ $17 < \ln \left[\varphi^3 (1 - \varepsilon_{mf}) \left(\frac{\rho_p}{\rho}\right) \text{Ar} \right] \leq 24$ <p>For Geldart Group D</p>			
Seo et al. (2014)	$\text{Re}_{mf} = 0.0005757 \text{Ar}$	135-385	2500	B
Tannous and Lourenço (2015)	$\text{Re}_{mf} = 0.0123 \text{Ar}^{0.8086}$	331-2017	395-2636	A, B, D

Reference Author(s)	Correlation	Particle diameter, d_p (μm)	Particle density, ρ_p (kg/m^3)	Geldart Group(s)
Gan et al. (2016)	$\text{Re}_{\text{mf}} = \frac{\varepsilon_{\text{mf}}^{3.041} d_p^{0.06} \tau^{0.298}}{78.89(1 - \varepsilon_{\text{mf}})} \text{Ar}$ Where τ = particle aspect ratio	25-10000	1349.5	A, B, D
Zarekar et al. (2016) 1	$\text{Re}_{\text{mf}} = 0.000711\text{Ar}$ $\text{Ar} \rightarrow 0$	120-728	2500	B, D
Zarekar et al. (2016) 2	$\text{Re}_{\text{mf}} = 0.1872\text{Ar}^{0.5}$ $\text{Ar} \rightarrow \infty$			

7.2.1 $\text{Re}_{\text{mf}} = A \cdot \text{Ar}^b$ type of correlations: Geldart Group A

This section evaluates all available $\text{Re}_{\text{mf}} = A \cdot \text{Ar}^b$ type of correlations for the same Geldart Group A particle properties listed in Table 7.2. Similar to the $\text{Re}_{\text{mf}} = (K_1^2 + K_2\text{Ar})^{0.5} - K_1$ type of correlations, d_p was chosen as the varying parameter because it is an important parameter in fluidization on which the Geldart classification is based.

Fig. 7.5. $\text{Re}_{mf} = A \cdot \text{Ar}^b$ type of correlations: Semi-log plot of U_{mf} vs. d_p for Geldart Group A

Fig. 7.5 displays the U_{mf} values predicted by the various correlations with respect to particle diameter (d_p) for Geldart Group A particles (Table 7.2). Comparing Fig. 7.5 and Fig. 7.2 (for $\text{Re}_{mf} = (K_1^2 + K_2 \text{Ar})^{0.5} - K_1$ correlations), three noteworthy observations were made.

Firstly, in both Fig. 7.5 and Fig. 7.2, U_{mf} increases with d_p and the rate of increase of U_{mf} with respect to d_p generally decreases as d_p increases. The Puspasari et al. (2013) D1 correlation is an oddity predicting an unphysical trend of U_{mf} decreasing with d_p when applied to all Geldart Groups A (Fig. 7.5), B (Fig. 7.6) and D (Fig. 7.7) particles, due to the empirical constant b having a value of 0.195 resulting from the oil palm leaves (low density and sphericity) investigated. Secondly, the discrepancy between the minimum and maximum U_{mf} values predicted is a few orders-of-magnitude greater than that for the previous type of correlation (Fig. 7.2), with the greatest discrepancy here being seven orders-of-magnitude. This can be attributed to the designation of the empirical constant b as an exponent of Ar in this type of correlations. Thirdly, in terms of orders-of magnitude, the greatest and least discrepancies are found at respectively the lowermost and uppermost d_p in Fig. 7.5, but is generally constant throughout the range of d_p in Fig. 7.2. This is presumably due to the varying empirical constant b as an exponent of Ar for the current type of correlation (Table 7.5), whereas that in the previous section has a fixed exponent (Table 7.1). Two other highlights related to the lower and higher U_{mf} predictions are described as follows. Regarding the lowest, both the correlations by Puspasari et al. (2013) for two different Geldart Group B particles (namely, B1 and B2) give the lowest U_{mf} values in the entire d_p range considered, which may be due to the significantly lower empirical constant A . It should be noted that applying these two correlations to Geldart Group B particles in Fig. 7.6 also results in outlying U_{mf} values, specifically for B1 in the highest U_{mf} values for d_p values larger than that investigated in the study ($d_p > 400 \mu m$) due to the empirical constant b being approximately 1.6. With respect to the higher U_{mf} predictions, four observations are described as follows. Firstly, the Puspasari et al. (2013) D1 correlation gives the highest U_{mf} predictions presumably due to the lower density and higher sphericity of the palm leaves investigated, since the other correlation for Geldart Group D (D2) does not give outlying U_{mf} values because it was developed from more regular Geldart Group D particles

(namely, glass spheres and food grains). Secondly, the Zarekar et al. (2016) 2 correlation predicts the second highest U_{mf} values, since it was theoretically formulated for hypothetically very large particles whereby Ar approaches infinity. Thirdly, the Subramani et al. (2007) 3, Jiliang et al. (2013) and Rabinovich and Kalman (2008) D correlations predict higher U_{mf} values presumably because the particles investigated are from Geldart Group B for the former two and Geldart Group D for the latter. However, when applied to the Geldart Group B test case (Fig. 7.6), the Subramani et al. (2007) 3 and Jiliang et al. (2013) correlations continue to over-predict U_{mf} presumably due to the very high value of empirical coefficient A (Table 7.6) and the consideration of particles at the boundary of Geldart Groups B and D, respectively; while the Rabinovich and Kalman (2008) D predicts reasonable U_{mf} values when applied to the Geldart Group D test case. Fourthly, both the correlations from the same authors (Thonglimp et al. (1984a) and Thonglimp et al. (1984b)) predict higher U_{mf} values due to the investigation of particles with Re_{mf} greater 20, thereby leading to discrepancies when applied to Geldart group A particles whose Re_{mf} values are less than 1.

7.2.2 $Re_{mf} = A \cdot Ar^b$ type of correlations: Geldart Group B

This section evaluates all available $Re_{mf} = A \cdot Ar^b$ type of correlations for Geldart group B particles, the properties of which are similar to that assessed in Section 7.1.2 and listed in Table 7.3.

Fig. 7.6 displays U_{mf} versus d_p for Geldart Group B particles. Compared to Fig. 7.5 for Geldart Group A particles, the span of U_{mf} values is smaller at between two and three orders-of-magnitude. As highlighted in previous reviews (Cahyadi et al., 2017; Cahyadi et al., 2015; Chew et al., 2015), the better accuracy could be due to the lack of inter-particle cohesive effects, which are not well understood, for the Geldart Group B particles here. A few interesting notes

are highlighted here. Firstly, the Gauthier et al. (1999) correlation gives low U_{mf} values because incipient fluidization velocity instead of complete fluidization velocity was evaluated for the particle mixtures (binary-size, Gaussian and wide PSDs). This indicates the need for bridging the gap between the monodisperse systems typically investigated (or rather, the lack of the reporting of the particle size distribution) and the more practical polydisperse systems. Secondly, as mentioned in the previous section, the Puspasari et al. (2013) B1 and B2 correlations give outlying predictions. While the B2 correlation yields low U_{mf} values throughout the Geldart Group B range, the B1 correlation yields the lowest U_{mf} values at the lowest d_p (90 μm) and the highest U_{mf} value at the highest d_p (630 μm). The behavior of the former can be attributed to low values of both empirical constants A and b , while that of the latter can be attributed to a small A but large b (~ 1.6). Thirdly, even though both the Rabinovich and Kalman (2008) B and Subramani et al. (2007) correlations were developed for Geldart group B particles, the former gives lower U_{mf} predictions while the latter higher U_{mf} predictions. This is due to the ad-hoc use of empirical constants to best-fit the experimental data – while the former assigned low value to the empirical constant A due to the low Reynolds numbers of the data considered (Bai et al., 1996; Chen et al., 1997; Formisani et al., 2002; Gauthier et al., 1999; Kim and Kim, 2002; Sanchez et al., 2003; Wu and Baeyens, 1998), the latter assigned a low power (-14.2) to ε_{mf} in the empirical constant A due to the non-consideration of the effect of temperature (300 – 975 K) on ε_{mf} in the correlation, which in turn affects the U_{mf} values (namely, ilmenite and quartz magnetite) assessed. Fourthly, the Mourad et al. (1994) correlation predicts higher U_{mf} values because it is based on Geldart Group D particles (namely, sand and corn). However, this correlation persists in giving higher U_{mf} values when applied to Geldart Group D particles in Fig. 7.7 presumably due to the very large (6.3 mm – 8 mm) corn particles used in this system. Finally, similar to that in Fig. 7.5 for Geldart Group A particles,

the Zarekar et al. (2016) correlation, which was developed for hypothetically high Ar ($Ar \rightarrow \infty$) systems, yields high U_{mf} values for Group B.

Fig. 7.6. $Re_{mf} = A \cdot Ar^b$ type of correlations: Semi-log plot of U_{mf} vs. d_p for Geldart Group B

7.2.3 $Re_{mf} = A \cdot Ar^b$ type of correlations: Geldart Group D

This section evaluates the $Re_{mf} = A \cdot Ar^b$ type of correlations (Table 7.5) for the same Geldart Group D particles evaluated earlier for the $Re_{mf} = (K_1^2 + K_2 Ar)^{0.5} - K_1$ type of correlation, the properties of which are listed in Table 7.4. Fig. 7.7 displays U_{mf} predictions with respect to d_p . Regarding the order-of-magnitude discrepancy between the maximum and minimum predictions, it is one at the lower d_p values and three at the higher d_p values, which is in contrast to Geldart Groups A (Fig. 7.5) and B (Fig. 7.6) whereby the spans of U_{mf} values are larger at the lower d_p values. The correlations giving higher U_{mf} predictions are assessed for the underlying reason for the inflation: (i) Puspasari et al. (2013) B1 has a higher empirical constant b , which also gives higher U_{mf} predictions for the larger Geldart Group B particles (Fig. 7.6); (ii) Maeno and Nishimura (1979)) has a higher values of empirical constants A and b , which can be attributed to the investigation of an unconventional particle system (namely, snow) and accentuates U_{mf} predictions for the Group D particles; (iii) Mourad et al. (1994) investigated a Geldart Group D binary mixture of sand and corn kernel, which thereby suggests the results from a binary mixture may not be directly applicable to monodisperse systems and underscores the need to specify particle size distributions; and (iv) Xie and Geldart (1995) investigated Geldart Group A particles, wherein inter-particle cohesion is dominant and thereby the U_{mf} values are inflated. On the other hand, the lower U_{mf} predictions are due to (i) the investigation of oil palm frond particles which are highly non-spherical and fibrous (i.e., low density) (Puspasari et al., 2013); and (ii) the investigation at elevated temperature (Jiliang et al. (2013)).

Fig. 7.7. $\text{Re}_{mf} = A \cdot \text{Ar}^b$ type of correlations: Semi-log plot of U_{mf} vs. d_p for Geldart Group D

7.2.4 Summary

For the $\text{Re}_{mf} = A \cdot \text{Ar}^b$ type of correlations, the following points can be summarized:

- (i) the shapes of U_{mf} versus d_p plots are dissimilar for some correlations, which contrasts to the similar shapes for the $\text{Re}_{mf} = (K_1^2 + K_2 \text{Ar})^{0.5} - K_1$ type of correlations; (ii) except for one

correlation [Puspasari et al. (2013) D1, which has a lower value of the exponent b resulting from low-density and sphericity particles], all others predict an increasing U_{mf} trend with respect to d_p ; (iii) the discrepancies among the U_{mf} predictions are greater than that for the $Re_{mf} = (K_1^2 + K_2 Ar)^{0.5} - K_1$ type of correlations, due to the presence of an empirical constant as an exponent; (iv) the ad-hoc inclusion or exclusion of the difficult-to-quantify parameters, ε_{mf} and φ , in this type of correlations contributes to larger discrepancies among predictions; (v) the greatest discrepancies among U_{mf} predictions are found for Geldart Group A particles, which agrees with that for the $Re_{mf} = (K_1^2 + K_2 Ar)^{0.5} - K_1$ type of correlations; and (vi) the smallest discrepancies are found for Geldart Group B predictions likely since this Group is the most studied type of particles for developing these type of U_{mf} correlations.

7.3 $Re_{mf} = \frac{Ar}{p+q\sqrt{Ar}}$ type of correlations

Only six correlations are available in this category. In this type of correlations, Re_{mf} is a function of both Ar and $\sqrt{Ar}$. Table 7.6 lists all the correlations in this category. The d_p , ρ_p and Geldart Group details are not reported for the first three correlations, while the Fletcher et al. (1992) correlation was based on Geldart Group A, B and D particles with $d_p = 75\text{-}3350\ \mu\text{m}$ and $\rho_p \approx 2500\ \text{kg/m}^3$ from past studies.

Table 7.6. $Re_{mf} = \frac{Ar}{p+q\sqrt{Ar}}$ type of correlations

Reference Author(s)	Correlation
Todes et al. (1958)	$Re_{mf} = \frac{Ar}{1400 + 5.2\sqrt{Ar}}$
Goroshkov et al. (1966)	$Re_{mf} = \frac{Ar}{150 \left(\frac{1 - \varepsilon_{mf}}{\varepsilon_{mf}^3} \right) + 1.323 \varepsilon_{mf}^{-1.5} \sqrt{Ar}}$
Davtyan et al. (1976)	$Re_{mf} = \frac{Ar}{1040 + 4.86\sqrt{Ar}}$
Fletcher et al. (1992) 1	$Re_{mf} = \frac{Ar}{1400 + 5\sqrt{Ar}}$ $\varphi > 0.9$
Fletcher et al. (1992) 2	$Re_{mf} = \frac{Ar}{1170 + \sqrt{Ar}}$ $0.8 < \varphi < 0.9$
Fletcher et al. (1992) 3	$Re_{mf} = \frac{Ar}{790 + 7\sqrt{Ar}}$ $0.8 < \varphi$

This section evaluates the six available $Re_{mf} = \frac{Ar}{p+q\sqrt{Ar}}$ type of correlations for Geldart Groups A, B and D particles, the base case parameters of which are listed in Tables 7.2, 7.3 and 7.4, respectively. Similarly, the U_{mf} values predicted are compared against varying particle diameters (d_p) while keeping all other parameters constant.

7.3.1 $Re_{mf} = \frac{Ar}{p+q\sqrt{Ar}}$ type of correlations: Geldart Groups A, B and D

Fig. 7.8 presents the plot of U_{mf} calculated by the various correlations against particle diameter (d_p) for Geldart Group A particles. Note that a logarithmic scale is not used for Figs. 7.8, 7.9 and 7.10, since the difference between maximum and minimum predictions is less than an order-of-magnitude. Although the various correlations are applied to Geldart Group A

systems, the trends in Fig. 7.8 are similar to Fig. 7.2 (on a non-logarithmic scale) but dissimilar to Fig. 7.5. Nonetheless, the underlying reason for the highest U_{mf} values predicted are due to the least spherical particles and different Geldart Groups (namely, B and D) investigated by Fletcher et al. (1992) for angular sands with particle sphericity (ϕ) of less than 0.8.

Fig. 7.8. $\text{Re}_{mf} = \frac{\text{Ar}}{p+q\sqrt{\text{Ar}}}$ type of correlations: U_{mf} vs. d_p for Geldart Group A

Fig. 7.9 displays the plot of U_{mf} calculated by the various correlations against particle diameter (d_p) for Geldart Group B particles. Similar to the observation for Geldart Group A particles, when the correlations are tested for Geldart Group B particles, the trends in Fig. 7.9 are similar to those in Fig. 7.3 (on a non-logarithmic scale). but not to those in Fig. 7.6. The

more significant outlying U_{mf} predictions in Fig. 7.9 are observed when $d_p > 400 \mu\text{m}$ by the Fletcher et al. (1992) correlation developed for sands with intermediate particle sphericity (namely, $0.8 < \phi < 0.9$), which matches the test case here (Table 7.3). The over-prediction can again be attributed to the ad-hoc empirical constants obtained through data-fitting, which limits the general applicability of the correlation.

Fig. 7.9. $\text{Re}_{mf} = \frac{\text{Ar}}{p+q\sqrt{\text{Ar}}}$ type of correlations: U_{mf} vs. d_p for Geldart Group B

Fig. 7.10 contains the plot of U_{mf} predicted against particle diameter (d_p) for Geldart Group D particles. In contrast to that for Geldart Groups A and B, the general trends of U_{mf} versus d_p for all three types of correlations in Figs. 7.4 (on a non-logarithmic scale), 7.7 and 7.10 are similar. Significantly higher U_{mf} predictions are clearly observed in Fig. 7.10 by the Fletcher et al. (1992) correlation developed for sands of intermediate particle sphericity (i.e., $0.8 < \phi < 0.9$), which also predicts higher U_{mf} values for the larger Geldart Group B particles in Fig. 7.9.

Fig. 7.10. $\text{Re}_{mf} = \frac{\text{Ar}}{p+q\sqrt{\text{Ar}}}$ type of correlations: U_{mf} vs. d_p for Geldart Group D

7.3.2 Summary

The $Re_{mf} = \frac{Ar}{p+q\sqrt{Ar}}$ type of correlation gives rather agreeable U_{mf} predictions, but significantly fewer correlations are of this type.

7.4 $U_{mf} = K X^\alpha$ type of correlations

The lump parameter X is a function of particle diameter (d_p), gas and particle densities (ρ_p and ρ), gas viscosity (μ) and gravitational acceleration (g). The exponents of the constituents of X were determined by dimensionless analysis, while the empirical constants K and α were obtained by data-fitting (Coltters and Rivas, 2004). Thus, the general format of this type of correlations is:

$$U_{mf} = K X^\alpha, \text{ where } X = \left(\frac{d_p^2(\rho_p - \rho)g}{\mu} \left(\frac{\rho_p}{\rho} \right)^{1.23} \right) \quad (7.10)$$

There are 15 correlations in a single publication (Coltters and Rivas, 2004) developed for different kinds of materials; these have been labelled as CR1 through CR15 in Table 7.7. Only three other papers have adopted this type of correlation. Table 7.7 lists the values of the empirical constants K and α reported.

Table 7.7. $U_{mf} = K X^\alpha$ correlations

Author(s)	K	α	Particle diameter, d_p (μm)	Particle density, ρ_p (kg/m^3)	Geldart Group(s)	Label	Remarks
Coltters and Rivas (2004)	4.7673×10^{-6}	0.71635	3-900	2700-11370	A, B, D	CR1	For metals
Coltters and Rivas (2004)	2.7568×10^{-6}	0.81455		768-2800		CR 2	For low- to mid-density alumina
Coltters and Rivas (2004)	3.7774×10^{-5}	0.6301		3300-4015		CR 3	For high-density alumina
Coltters and Rivas (2004)	4.3384×10^{-7}	0.89029	23-569		A, B	CR 4	For glass
Coltters and Rivas (2004)	2.4624×10^{-3}	0.46943	569-3000		B, D	CR 5	
Coltters and Rivas (2004)	9.7119×10^{-7}	0.84268	95-800		B, D	CR 6	For sand
Coltters and Rivas (2004)	6.4051×10^{-3}	0.42520	800-2800		D	CR 7	
Coltters and Rivas (2004)	4.7731×10^{-6}	0.87117	710-1000		B, D	CR 8	For coal
Coltters and Rivas (2004)	8.5557×10^{-3}	0.46093	1000-3578		D	CR 9	
Coltters and Rivas (2004)	1.145×10^{-5}	0.71957	25-2250	790-2950	A, B, D	CR 10	For catalysts

Author(s)	K	α	Particle diameter, d_p (μm)	Particle density, ρ_p (kg/m^3)	Geldart Group(s)	Label	Remarks
Coltters and Rivas (2004)	3.1108×10^{-8}	0.93283	101-1250			CR 11	For metallic ores
Coltters and Rivas (2004)	2.1308×10^{-4}	0.59460	116-1000			CR 12	For polymers
Coltters and Rivas (2004)	4.427×10^{-3}	0.47851	502-2828			CR 13	For orthorhombic crystals
Coltters and Rivas (2004)	7.9265×10^{-4}	0.50953	0.89-2300			CR 14	For hexagonal crystals
Coltters and Rivas (2004)	7.1187×10^{-5}	0.61787	106-2474			CR 15	For cubic crystals
Zhong et al. (2008)	1.45×10^{-3}	0.363	500-6500	274-2700	B, D		
Oliveira et al. (2013)	1.17×10^{-4}	0.4916	125-1400	1302-2696	B, D		
Shao et al. (2013)	1.28×10^{-3}	0.356	180-10000	677-2560	B, D		

7.4.1 $U_{mf} = K X^\alpha$ type of correlations: Geldart Group A

This section applies the correlations listed in Table 7.7 to Geldart Group A particles, whose properties are listed in Table 7.2. Fig. 7.11 displays the plot of U_{mf} calculated by the various correlations against particle diameter (d_p) for Geldart Group A particles. The trends are similar to that for the $\text{Re}_{mf} = A \cdot \text{Ar}^b$ type of correlations in Fig. 7.5. Five orders-of-magnitude of discrepancy exists between the minimum and maximum U_{mf} predictions. The lowest U_{mf} predictions is due to the lowest K value (CR11), which may be due to the low particle sphericity

of the metallic oxide investigated. The second and third lowest U_{mf} predictions are from CR4 and CR6, respectively, which can again be attributed to the lower K values. However, these correlations cease to be outliers when applied to Geldart Groups B ($d_p > 200 \mu\text{m}$) and D particles due to their relatively high α values (> 0.8). Note that CR4 was developed for Geldart Groups A and B particles, while CR6 for Geldart Groups B and D particles. On the other hand, the higher U_{mf} predictions are due to: (i) a different Geldart Group used to develop the correlation, specifically Geldart Groups B (CR5) and D (CR7, CR9, CR12-15); (ii) different particle shapes, specifically, orthorhombic (CR13), hexagonal (CR14), cubic (CR15) and needle-like (Zhong et al.); and (iii) different particle density, specifically, alumina, whose particle density is much higher (CR3). The correlations predicting high U_{mf} values tend to have higher K values and lower α values.

Fig. 7.11. $U_{mf} = K X^\alpha$ type of correlations: Semi-log plot of U_{mf} vs. d_p for Geldart Group A

7.4.2 $U_{mf} = K X^\alpha$ type of correlations: Geldart Group B

This section evaluates the available $U_{mf} = K X^\alpha$ type of correlations for Geldart Group B particles, whose properties are listed in Table 7.3. Fig. 7.12 plots U_{mf} versus particle diameter (d_p) for the Geldart Group B particles. The predictions span up to three orders-of-magnitude. Similar to that in Fig. 7.11 for Geldart Group A particles, the CR11 correlation gives

significantly lower U_{mf} values due to the lowest K value. In addition, although the Oliveira et al. (2013) correlation was developed for Geldart Group B particles, the U_{mf} predictions are among the lowest, presumably due to the low particle densities and sphericities of the biomass particles used. Also similar to that in Fig. 7.11 for Geldart Group A particles, high values of the system-specific empirical constants K (CR9, CR13, CR14, CR5, CR7 and CR12) and α (CR8) lead to higher U_{mf} predictions.

Fig. 7.12. $U_{mf} = K X^\alpha$ type of correlations: Semi-log plot of U_{mf} vs. d_p for Geldart Group B

7.4.3 $U_{mf} = K X^\alpha$ type of correlations: Geldart Group D

This section evaluates the $U_{mf} = K X^\alpha$ type of correlations (Table 7.4) for Geldart Group D particles (Table 7.4). Fig. 7.13 plots U_{mf} predicted against particle diameter (d_p) for Geldart Group D particles. The span of discrepancy between the minimum and maximum U_{mf} predictions is three orders-of-magnitude. Similar to that for Geldart Groups A and B, the CR11 correlation persists in giving the lowest U_{mf} predictions for Geldart Group D particles. The under-prediction across all Geldart Groups indicates that particle sphericity is a dominant parameter in affecting U_{mf} , thereby will be looked at in more detail in the next section. Two other correlations that give low U_{mf} predictions are that by Shao et al., 2013 and Oliveira et al, 2013. While Shao et al., 2013 investigated particle mixtures involving Geldart Groups C, B and D, Oliveira et al, 2013 investigated Geldart Group B particles. This is consistent with the observations so far that polydisperse particle mixtures and a different Geldart Group used to develop the correlations tend to give outlying U_{mf} values. Regarding the high U_{mf} predictions, (i) despite being developed for Geldart Group D particles, the CR8 and CR9 correlations give higher U_{mf} values, presumably because of the specific particle type (namely, coal) and the fact that the correlations have been developed only from six data-points each; (ii) the higher values of the empirical coefficient K , leads to higher U_{mf} predictions by the CR13, CR14, CR5, CR7 and CR12 correlations, presumably due to skewed datasets because of the higher d_p limit ($\sim 2000 \mu\text{m}$) used in the development of these correlations.

Fig. 7.13. $U_{mf} = K X^\alpha$ type of correlations: Semi-log plot of U_{mf} vs. d_p for Geldart Group D

7.4.4 Summary

The following conclusions can be drawn from the comparative analysis of the U_{mf} predictions from the $U_{mf} = K X^\alpha$ type of correlations for Geldart Groups A, B and D: (i) the shapes of the U_{mf} trends are not as similar as those for the $\text{Re}_{mf} = (K_1^2 + K_2 \text{Ar})^{0.5} - K_1$ type of correlations, but not as dissimilar as those for the $\text{Re}_{mf} = A \cdot \text{Ar}^b$ type of correlations; (ii) all correlations predict an increase in U_{mf} with d_p ; (iii) the greatest discrepancy among U_{mf}

predictions is four orders-of-magnitude, due to the presence of an empirical exponent per that for the $Re_{mf} = A \cdot Ar^b$ type; (iv) correlations developed from less conventional particle systems (e.g. hexagonal crystals, metallic ores etc.) increase discrepancies among predictions, which implies the importance of properly quantifying and reporting particle properties; (v) similar to correlations of types $Re_{mf} = (K_1^2 + K_2 Ar)^{0.5} - K_1$ and $Re_{mf} = A \cdot Ar^b$, predictions for Geldart Group A particles exhibit the greatest discrepancies; and (vi) unlike correlations of types $Re_{mf} = (K_1^2 + K_2 Ar)^{0.5} - K_1$ and $Re_{mf} = A \cdot Ar^b$, predictions for Geldart Groups B and D show a similar order-of-magnitude of discrepancies, presumably due to the scrutiny of relatively more Geldart Group D particles in the development of this type of correlations.

7.5 Correlations involving voidage (ε_{mf}) and sphericity (φ)

The bed voidage at minimum fluidization (ε_{mf}) and particle sphericity (φ) values are not typically available, these are usually subsumed into empirical constants via best-fitting of experimental data. To understand the effect of ε_{mf} and φ on U_{mf} predictions, this section compiles all correlations involving ε_{mf} and φ in Table 7.8.

Table 7.8. Correlations involving both ε_{mf} and φ

Correlation	Type	Reference
$Re_{mf} = \left(\frac{\varphi^2 \varepsilon_{mf}^5}{18 + 9(1 - \varepsilon_{mf})} \right) Ar, Re_{mf} < 2$	$Re_{mf} = A \cdot Ar^b$	Johnson (1949-50)
$Re_{mf} = \left(\left(\frac{42.86(1 - \varepsilon_{mf})}{\varphi} \right)^2 + 0.571 \varepsilon_{mf}^3 \varphi Ar \right)^{0.5} - \left(\frac{42.86(1 - \varepsilon_{mf})}{\varphi} \right)$	$Re_{mf} = (K_1^2 + K_2 Ar)^{0.5} - K_1$	Ergun (1952)

Correlation	Type	Reference
$\text{Re}_{\text{mf}} = \frac{\text{Ar}}{150 \left(\frac{1 - \varepsilon_{\text{mf}}}{\varepsilon_{\text{mf}}^3} \right) + 1.323 \varepsilon_{\text{mf}}^{-1.5} \sqrt{\text{Ar}}}$	$\text{Re}_{\text{mf}} = \frac{\text{Ar}}{p + q\sqrt{\text{Ar}}}$	Goroshkov et al. (1966)
$\text{Re}_{\text{mf}} = \frac{\varepsilon_{\text{mf}}^3}{150(1 - \varepsilon_{\text{mf}})} \text{Ar}$	$\text{Re}_{\text{mf}} = A \cdot \text{Ar}^b$	Maeno and Nishimura (1979)
$\text{Re}_{\text{mf}} = \left([83.33(1 - \varepsilon_{\text{mf}})]^2 + 0.417 \varepsilon_{\text{mf}}^3 \text{Ar} \right)^{0.5} - [83.33(1 - \varepsilon_{\text{mf}})]$	$\text{Re}_{\text{mf}} = (K_1^2 + K_2 \text{Ar})^{0.5} - K_1$	Pattipati and Wen (1981)
$\text{Re}_{\text{mf}} = \left[\left(33.3 \frac{1 - \varepsilon_{\text{mf}}}{\varphi} \right)^2 + 0.575 \varphi \varepsilon_{\text{mf}}^3 \text{Ar} \right]^{0.5} - \left(33.3 \frac{1 - \varepsilon_{\text{mf}}}{\varphi} \right)$	$\text{Re}_{\text{mf}} = (K_1^2 + K_2 \text{Ar})^{0.5} - K_1$	Tannous et al. (1994)
$\text{Re}_{\text{mf}} = \frac{\varepsilon_{\text{mf}}^3}{180(1 - \varepsilon_{\text{mf}})} \text{Ar}$	$\text{Re}_{\text{mf}} = A \cdot \text{Ar}^b$	Xie and Geldart (1995)
$\text{Re}_{\text{mf}} = \left((600 \varepsilon_{\text{mf}}^3 (1 - \varepsilon_{\text{mf}}))^2 + 0.0408 \text{Ar} \right)^{0.5} - (600 \varepsilon_{\text{mf}}^3 (1 - \varepsilon_{\text{mf}}))$	$\text{Re}_{\text{mf}} = (K_1^2 + K_2 \text{Ar})^{0.5} - K_1$	Delebarre (2004)
$\text{Re}_{\text{mf}} = \left(\frac{0.3507}{\varepsilon_{\text{mf}}} \right)^{14.2} \text{Ar}^{0.55}$ Temperature: 250-900 K	$\text{Re}_{\text{mf}} = A \cdot \text{Ar}^b$	Subramani et al. (2007)
$\text{Re}_{\text{mf}} = 0.2029 \left(\frac{\varphi}{\varepsilon_{\text{mf}}} \right)^{0.7} \left(\frac{\rho}{\rho_p - \rho} \right)^{0.5} \text{Ar}^{0.75}$	$\text{Re}_{\text{mf}} = A \cdot \text{Ar}^b$	de Vasconcelos and Mesquita (2011)
$\text{Re}_{\text{mf}} = 3.92 \cdot 10^{-6} \varphi^{1.826} (1 - \varepsilon_{\text{mf}})^{0.942} \left(\frac{\rho_p}{\rho} \right)^{0.942} \text{Ar}^{0.942}$ $6 \leq \ln \left[\varphi^3 (1 - \varepsilon_{\text{mf}}) \left(\frac{\rho_p}{\rho} \right) \text{Ar} \right] \leq 9$ For Geldart Group A	$\text{Re}_{\text{mf}} = A \cdot \text{Ar}^b$	Puspasari et al. (2013) A
$\text{Re}_{\text{mf}} = 7.66 \cdot 10^{-10} \varphi^{3.734} (1 - \varepsilon_{\text{mf}})^{1.578} \left(\frac{\rho_p}{\rho} \right)^{1.578} \text{Ar}^{1.578}$ $12.5 \leq \ln \left[\varphi^3 (1 - \varepsilon_{\text{mf}}) \left(\frac{\rho_p}{\rho} \right) \text{Ar} \right] \leq 15$ For Geldart Group B	$\text{Re}_{\text{mf}} = A \cdot \text{Ar}^b$	Puspasari et al. (2013) B1

Correlation	Type	Reference
$\text{Re}_{mf} = 7.02 \cdot 10^{-7} \varphi^{1.958} (1 - \varepsilon_{mf})^{0.986} \left(\frac{\rho_p}{\rho} \right)^{0.986} \text{Ar}^{0.986}$ $15 < \ln \left[\varphi^3 (1 - \varepsilon_{mf}) \left(\frac{\rho_p}{\rho} \right) \text{Ar} \right] \leq 16.5$ For Geldart Group B	$\text{Re}_{mf} = A \cdot \text{Ar}^b$	Puspasari et al. (2013) B2
$\text{Re}_{mf} = 2.42 \varphi^{-0.415} (1 - \varepsilon_{mf})^{0.195} \left(\frac{\rho_p}{\rho} \right)^{0.195} \text{Ar}^{0.195}$ $15 \leq \ln \left[\varphi^3 (1 - \varepsilon_{mf}) \left(\frac{\rho_p}{\rho} \right) \text{Ar} \right] \leq 17$ For Geldart Group D	$\text{Re}_{mf} = A \cdot \text{Ar}^b$	Puspasari et al. (2013) D1
$\text{Re}_{mf} = 6.96 \cdot 10^{-4} \varphi^{0.77} (1 - \varepsilon_{mf})^{0.59} \left(\frac{\rho_p}{\rho} \right)^{0.59} \text{Ar}^{0.59}$ $17 < \ln \left[\varphi^3 (1 - \varepsilon_{mf}) \left(\frac{\rho_p}{\rho} \right) \text{Ar} \right] \leq 24$ For Geldart Group D	$\text{Re}_{mf} = A \cdot \text{Ar}^b$	Puspasari et al. (2013) D2
$\text{Re}_{mf} = ((30.81 \varphi^2)^2 + 0.0674 \text{Ar})^{0.5} - 30.81 \varphi^2$	$\text{Re}_{mf} = (K_1^2 + K_2 \text{Ar})^{0.5} - K_1$	Kumoro et al. (2014)
$\text{Re}_{mf} = \frac{\varepsilon_{mf}^{3.041} d_p^{0.06} \tau^{0.298}}{78.89(1 - \varepsilon_{mf})} \text{Ar}$ Where τ = particle aspect ratio	$\text{Re}_{mf} = A \cdot \text{Ar}^b$	Gan et al. (2016)
$\text{Re}_{mf} = \left(\left(\frac{25.7}{\varphi} \right)^2 + 0.0365 \varphi \text{Ar} \right)^{0.5} - \frac{25.7}{\varphi}$ (no slip condition)	$\text{Re}_{mf} = (K_1^2 + K_2 \text{Ar})^{0.5} - K_1$	Zarekar et al. (2016)

7.5.1 Prediction of minimum fluidization velocity with respect to bed voidage at minimum fluidization

This section evaluates all available correlations involving ε_{mf} (Table 7.8) for Geldart Groups A, B and D particles, whose particle properties are listed in Tables 7.2, 7.3 and 7.4, respectively. It should be noted that the specific ε_{mf} values listed in Tables 7.2, 7.3 and 7.4 will not be used, as ε_{mf} is varied between 0.3 and 0.7 in this section to assess the impact of the ε_{mf}

value. Also, d_p values for particles of Geldart Groups A, B and D will be kept constant at 90 μm , 360 μm and 1600 μm respectively.

Fig. 7.14 displays the U_{mf} values calculated by the various correlations against voidage at minimum fluidization (ε_{mf}), with each of the three sub-plots representing Geldart Groups A, B and D. Five observations are worth noting. Firstly, among the correlations, the predicted U_{mf} values span up to five orders-of-magnitude for all three Groups. However, removing one correlation, namely, the one by Subramani et al. (2007), reduces the discrepancy to about three orders-of-magnitude for Geldart Groups B and D (Fig. 7.14b and c). Secondly, across the Geldart Groups, the trends are similar for every correlation. Thirdly, some correlations predict that U_{mf} increases with ε_{mf} while others predict a decrease, which highlights a need for a better understanding of ε_{mf} . The positive and negative relationships between U_{mf} and ε_{mf} is due to the respectively positive and negative exponents of ε_{mf} in the correlations. Those correlations which predict an increase of U_{mf} with ε_{mf} show a change of between two and three orders-of-magnitude in the range of ε_{mf} investigated, whereas those correlations which predict a decrease of U_{mf} with ε_{mf} (with the exception of Subramani et al. (2007)) exhibit a smaller change of less than an order-of-magnitude. It is interesting to note that the former type of correlations are of the type $\text{Re}_{mf} = (K_1^2 + K_2 \text{Ar})^{0.5} - K_1$. Fourthly, the most significant change of U_{mf} values in the range of ε_{mf} evaluated here is about five orders-of-magnitude by Subramani et al. (2007), because the ε_{mf} values of between 0.46 and 0.54 investigated led to a correlation whereby U_{mf} is proportional to $\varepsilon_{mf}^{-14.2}$ (Table 7.8), the large negative exponent of which made U_{mf} extremely sensitive to ε_{mf} . Fifthly, the smallest change of about 1.2-fold of U_{mf} values with ε_{mf} is by the Puspasari et al. (2013) D1 correlation, whereby U_{mf} is proportional to $(1 - \varepsilon_{mf})^{0.195}$. This underscores the importance of correctly accounting for ε_{mf} when developing correlations.

Fig. 7.14. Semi-log plot of U_{mf} versus ε_{mf} for (a) Geldart Group A, (b) Geldart Group B and (c) Geldart Group D particles.

7.5.2 Prediction of minimum fluidization velocity with respect to particle sphericity

This section evaluates the available correlations involving ϕ (Table 7.8) for Geldart Groups A, B and D particles, the particle properties of which are listed in Tables 7.2, 7.3 and 7.4, respectively. The particle sphericity (ϕ) parameter does not adhere to the values in the Tables since the effect of its variation between 0.3 and 1 is of interest here. Also, d_p values for particles of Geldart Groups A, B and D are kept constant at 90 μm , 360 μm and 1600 μm , respectively.

Fig. 7.15a, b and c display the plot of U_{mf} calculated by the various correlations against particle sphericity (ϕ) for Geldart Groups A, B and D, respectively. Four noteworthy observations are described as follows. Firstly, a discrepancy of about six orders-of-magnitude is clear between the maximum and minimum U_{mf} predictions for Geldart Group A particles,

while that for Geldart Groups B and D particles is only about three orders-of-magnitude. This may be attributed to the lack of understanding and thereby accounting of inter-particle cohesive effects unique to Geldart Group A. Secondly, the trends for each correlation are generally similar across all Geldart Groups, which is similar to that in Fig. 7.14. Thirdly, only the Puspasari et al. (2013) D1 and Kumoro et al. (2014) correlations predict a decrease in U_{mf} with increasing ϕ , while the rest predict an increase in U_{mf} with ϕ . The predicted decrease in U_{mf} with increasing ϕ is due to the negative exponent of ϕ in the Puspasari et al. (2013) D1 correlation, while that in the Kumoro et al. (2014) correlation is due to the negative term of $-30.81\phi^2$, which increases as ϕ increases. Fourthly, the change in U_{mf} values in the range of ϕ values (i.e., 0.3 - 1) is up to two orders-of-magnitude lesser than that in the range of ε_{mf} values (i.e., 0.3 - 0.7). This could be associated with the range of ε_{mf} and ϕ values considered, with a wider range physically possible for the latter. The most significant variation of U_{mf} of about two orders-of-magnitude in the ϕ range evaluated is by the Puspasari et al. (2013) B1 correlation due to a more positive exponent (i.e., 3.7) of ϕ , while the smallest variation of about 1.6-fold is by the Puspasari et al. (2013) D1 correlation due to a slightly negative exponent (i.e., -0.4) of ϕ .

Fig. 7.15. Prediction of U_{mf} with respect to ϕ : Semi-log plot of U_{mf} vs. ϕ for (a) Geldart Group A, (b) Geldart Group B and (c) Geldart Group D particles.

7.6 Discussion

In order to compare the spread of U_{mf} values across the four types of correlations and three Geldart Groups (namely A, B and D), the Geometric Standard Deviation (GSD) of the data is compared. Specifically, the GSD of each type of correlations at each d_p is defined as the anti-logarithm of the arithmetic standard deviation of the natural logarithms of the data, and calculated by:

$$\text{GSD} = \exp \left[\sqrt{\frac{\sum_{i=1}^n \{ \ln(U_{mf,i}) - \overline{\ln(U_{mf,i})} \}^2}{n}} \right] \quad (7.11)$$

where

$$\overline{\ln(U_{mf,i})} = \frac{\sum_{i=1}^n \ln(U_{mf,i})}{n} \quad (7.12)$$

and $U_{mf,i}$ is the U_{mf} predicted at a d_p by a correlation of the category for which GSD is being determined and n is the number of correlations in the category for which the GSD is being determined.

The GSD is adopted instead of the arithmetic standard deviation because the data is spread over multiple orders-of-magnitude. Fig. 7.16a shows the GSD of the U_{mf} values predicted by the four categories of correlations for the three Geldart Groups (i.e., A, B and D) versus d_p , while Fig. 7.16b shows that versus ε_{mf} and φ .

Three noteworthy inferences from Fig. 7.16a can be drawn: (i) omitting the $Re_{mf} = \frac{Ar}{p+q\sqrt{Ar}}$ type of correlation due to the limited number of correlations (i.e., six), the $Re_{mf} = (K_1^2 + K_2 Ar)^{0.5} - K_1$ type of correlation exhibits the least discrepancy among the U_{mf} predictions, which appears to be due to having a fixed exponent rather than an empirical constant as an exponent; (ii) the $U_{mf} = K X^\alpha$ type of correlations exhibit the greatest discrepancies among the U_{mf} predictions for $d_p > 40 \mu m$ presumably because they were developed from less conventional particle systems; (iii) the spread of U_{mf} predictions is generally greatest for Geldart Group A particles due to the lack of understanding and thereby accounting of inter-particle cohesive effects.

Fig. 7.16b reveals the following: (i) the maximum GSD values are greater in Fig. 7.16b than Fig. 7.16a, which indicate the sensitivity of the U_{mf} predictions to ε_{mf} and φ , and thereby the need for more understanding on such less readily available values; (ii) similar to Fig. 7.16a, Geldart Group A exhibits the greatest GSD values; and (iii) with respect to φ , GSD decreases as φ increases for Geldart Groups A and B, but increases with φ for Geldart Group D; (iv) with respect to ε_{mf} , GSD decreases then increases with ε_{mf} for all Geldart Groups, with the minima between $\varepsilon_{mf} = 0.4$ and $\varepsilon_{mf} = 0.5$, because the correlations are largely based on this range of ε_{mf} .

Fig. 7.16. Geometric Standard Deviation (GSD) of predicted U_{mf} values versus (a) d_p ; and (b) ε_{mf} and ϕ .

7.7 Conclusion

A comprehensive review of all the correlations available in literature to date for determining the minimum fluidization velocity (U_{mf}) has been conducted in this study. The correlations are classified into four types and a comparison of correlations in each category is carried out for three Geldart Groups, namely, A, B and D. Furthermore, the sensitivity of the correlations to the less readily available terms of voidage at minimum fluidization (ε_{mf}) and particle sphericity (ϕ) are also assessed.

The key conclusion is that the correlations available in literature for predicting U_{mf} are highly empirical and system-specific, which severely limit the utility of each to systems beyond the scope tested. A mechanistically based correlation may be still intractable at this point, so the following recommendations are made for future studies on improving the prediction of U_{mf} . Firstly, when formulating new correlations, the use of empirical coefficients as exponents is particularly detrimental when the correlation is applied to systems beyond the specific scope investigated. Secondly, the extent of inter-particle interactions should be accounted for when formulating U_{mf} correlations, particularly for application to Geldart Group A particles. This agrees with previous observations regarding transport disengaging height (TDH) (Cahyadi et al., 2015) and entrainment (Chew et al., 2015), whereby the lack of understanding of inter-particle cohesive and clustering effects also led to orders-of-magnitudes of discrepancies in the predictions. Thirdly, the less readily available values of ε_{mf} and ϕ needs more attention, in view of the sensitivity of the U_{mf} predictions to these parameters. Fourthly, more comprehensive and detailed reporting of the following are needed to allow for advancing the knowledge base involving U_{mf} : (i) particle properties, particularly the particle size distribution, and (ii) distributor and reactor configurations.

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8

Literature review: Circulating fluidized beds (CFBs)

This chapter provides a comprehensive literature survey of the research works on CFBs particularly in context of pressure signal analysis and the core-annulus solids flow structure.

8.1 Pressure signal analysis in CFBs

Pressure transmitters are the most common instrumentation in commercial fluidized beds, due to the ease of measurement even under harsh conditions, and also to their robustness, low-cost and non-intrusive nature (which allows for measurements to be taken without distorting the flow) (Tahmasebpour et al., 2013; van Ommen et al., 2011). Another instrumentation available for routine measurement is for temperature (Werther, 1999). However, the temperature signal features a dynamic much slower than the pressure signal dynamic, since the latter is based on pneumatics. Hence, the data obtained from pressure transmitters are often the only useful data available for understanding the hydrodynamics in fluidized beds. This necessitates efforts to extract as much valuable information from pressure signals as possible.

Unsurprisingly, studies on using pressure signals to characterize the dynamics of gas-solid fluidized beds are not lacking (Johnsson et al., 2000; Sasic et al., 2007). Fluctuations in the pressure signals have been used to identify fluidization regimes (Bi et al., 2000; Felipe and Rocha, 2007; Johnsson et al., 2000; Sobrino et al., 2008; Yerushalmi and Cankurt, 1979), analyze defluidization (Briens et al., 2003; Parise et al., 2009; van Ommen et al., 2004), predict average bubble sizes (van der Schaaf et al., 2002; Zhang et al., 2010), detect agglomeration (Bartels et al., 2010; Chirone et al., 2006; van Ommen et al., 1999), measure moisture content

(Chaplin et al., 2004), and indicate heterogeneity (Gheorghiu and Coppens, 2004). The amplitudes of ΔP signals have been used to provide the ‘fluidization quality index’, which was defined as the ratio of the average amplitude of ΔP to its dominant frequency (Bi, 2007; Shuster and Kisliak, 1952). Also, the standard deviation of pressure signals has been used to identify the ideal fluidization velocity (Chen et al., 2006). Studies on the propagation of pressure waves (Bi et al., 1995; Musmarra et al., 1995; Roy et al., 1990; Ryzhkov and Tolmachev, 1983; Tallon and Davies, 1999) and their impact on pressure measurement at a specific location have also been carried out (Bi, 2007).

Pressure signals can be analyzed with time-series analysis of linear (i.e., time, frequency, or time-frequency domain) or non-linear (i.e., state space) methods (Bi and Li, 2004; Li and Kwauk, 2003; van Ommen et al., 2011). Such methods aim to dissect the signal components and study the phenomena contributing to each of them. Indirect and statistical analysis in the time domain (e.g., standard deviation, probability density function, autoregressive function, or Hurst analysis method) is a simple, signal amplitude-based method which ignores time scale of the signals (van Ommen et al., 2011) (except the average time cycle method (Briens and Briens, 2002)). These methods have been applied to detect agglomeration (Bartels et al., 2010; van Ommen et al., 1999), regime transitions (Johnsson et al., 2000; van Ommen et al., 2011), gas bypassing (Issangya et al., 2010; Issangya et al., 2013), and variations in superficial gas velocity, bed density, segregation (Chaplin et al., 2004; Fan et al., 1981; van Ommen et al., 1999), and also for online monitoring in industry (van Ommen et al., 2011). Frequency methods (e.g., power spectrum, transient power spectral density) are largely based on Fourier transform (van Ommen et al., 2011). Power spectrum analysis has been used to determine fluidization regime changes (He et al., 2014), while dominant frequency analysis, narrow band standard deviation analysis and attractor comparison tool have been used to analyze pressure signals to determine granule moisture content and particle size (de Martín

et al., 2011). Both power spectrum and dominant frequency analysis of pressure signals have been used as indicators of similarity between laboratory and industry scaled fluidized beds for testing scale-up correlations (Glicksman et al., 1993; Nicastrò and Glicksman, 1984). Chaos or non-linear analysis is based on state space variables (i.e., correlation dimension, Kolmogorov entropy, Lyapunov exponents, etc.) which are rooted in the equations of motion of gas and particles and is more sensitive to variable changes (e.g., moisture content (Chaplin et al., 2004)) in fluidized bed phenomena than frequency analysis (Johnsson et al., 2000; van Ommen et al., 2011). Recently, a nonlinear time series analysis technique, recurrence quantification analysis, has been used to identify various structures in a fluidized bed (Tahmasebpour et al., 2015).

As reviewed above, studies on pressure signals in fluidized beds are wide-ranging. The focus here is on assessing differential pressure (ΔP) signals from a comprehensive dataset (involving 11 signals axially along the riser, five particle systems and four operating conditions) as a means of obtaining (i) a mechanistic understanding on the hydrodynamics (e.g., gas-particle interaction), and (ii) more in-depth information regarding the process. Fluidized bed systems are well acknowledged to exhibit wide over-lapping spectra of gas and particle concentrations that continuously change, thus the identification of fluidization characteristics warrants a data analysis method that retains the dynamic nature of the fluidization process (Chew et al., 2015; Chew et al., 2012a; Ellis et al., 2003; Guenther and Breault, 2007; Ren and Li, 1998; Yang and Leu, 2009). To this end, a simple averaging may be inadequate, which hence led to the use of Discrete Fourier Transform (DFT) (Drahoš et al., 1988; Fan et al., 1981; He et al., 2014; Jaiboon et al., 2013; Johnsson et al., 2000; Satija and Fan, 1985; Tahmasebpour et al., 2013; van der Schaaf et al., 1999) and wavelet decomposition (Chew et al., 2015; Chew et al., 2012a; Ellis et al., 2003; Guenther and Breault, 2007; Mallat, 1989, 2009; Ren and Li, 1998; Yang and Leu, 2009). On one hand, DFT has been widely used to identify regime changes in fluidized beds (Drahoš et al., 1988; He et al., 2014; Jaiboon et al., 2013; Johnsson

et al., 2000; Satija and Fan, 1985; Tahmasebpour et al., 2013; van der Schaaf et al., 1999). On the other hand, wavelet decomposition (Mallat, 1989, 2009) has been reported to be advantageous due to the ability to extract different frequency ranges while retaining the timestamp of signals, thereby facilitating the recognition of critical features (Chew et al., 2012a; Ellis et al., 2003; Ren and Li, 1998) of fluidized bed measurement data in real time.

8.2 Core-annulus flow in CFBs

The core-annulus phenomenon was first proposed by Bierl and Gajdos (1982). They proposed a flow structure, which consisted of a dilute core of upward moving particles, which occupied most of the cross-section, and a relatively dense annulus at the riser wall, which consisted mainly of downward-moving solids. Weinstein et al. (1986) also acknowledged the core-annulus structure for describing the radial solids distribution in a CFB. Dry (1986) used a technique based on the equivalence of heat and mass transfer to determine the radial solids profiles in a 7.2 m tall fast-fluidized bed and found that the core-annulus structure developed above a height of 0.4 m from the entrance zone. Johnsson et al. (2000) concluded that the concentration of solids in the annulus at the wall may be as high as the bulk solids density. Zhang et al. (1991) showed that the radial voidage profile became steeper with increasing solids circulation rate or decreasing gas velocity. This corresponded with the results of Monceaux et al. (1986) who had shown that the core-annulus flow was enhanced, i.e., the riser had greater solids upflow mass flux at the core and greater downflow solids mass flux at the annulus with increasing solids circulation rate. Herb et al. (1992) also observed a core-annulus profile in the riser but they found some downward solids flux in the core, which they attributed to occasional particle clusters, which moved downward due to their greater characteristic mass. They noted that the thickness of the annulus was approximately an order-of-magnitude lower than the column diameter, or column width in the case of a square riser, and that risers with different

diameters gave similar radial voidage profiles. Rhodes and Lausmann (1992) showed significant variation in the solids flux in the annulus along the perimeter of the riser, while Yang et al. (1993) also found greater variation of the local gas velocity in the annulus. Qi and Farag (1993) found higher solids flux near the wall, which exceeded the overall solids flux. Zhou et al. (1994) observed a core-annulus flow in a CFB riser of square cross-section and noted that the particle concentration was not always minimum at the central axis of the column. The 'M' shaped concentration profiles observed agreed with the voidage data for circular risers by Bai et al. (1992). Zhou et al. (1994) also found that the core had a more homogeneous flow than the wall. Several authors have noted that the radial solids concentration became more uniform with riser height (Brereton and Grace, 1993; Kato et al., 1991; Yang et al., 1991). In addition, coarser particles have been observed to preferentially segregate to the annulus (Bodelin et al., 1994; Chew et al., 2011; Dry, 1987). Bi and Grace (1995) were the first to include the core-annulus phenomenon as a separate regime in a regime map of riser flows involving Geldart Group A and smaller Geldart Group B particles, constructed using semi-empirical correlations published earlier (Bi and Fan, 1991; Leung, 1980; Yousfi and Gau, 1974).

Tsuo and Gidaspow (1990) predicted the core-annulus structure using multiphase flow equations to simulate a two-dimensional CFB riser. They obtained a general quantitative agreement between predicted and experimental values of solid velocity, with some discrepancies at the transition of the core and annulus. Yang et al. (2003) studied the core-annulus flow in a CFB riser via a CFD simulation using the energy minimization multi-scale (EMMS) approach and varying drag coefficients of different structures in the core and annulus. Outlet solid flux values and radial and axial voidage distributions calculated via the simulations were found to quantitatively agree with experimental results. Benyahia et al. (2000) employed a two-dimensional model using the kinetic theory of solid particles to simulate a CFB riser and

predicted a core-annulus flow from solid flux and granular temperature profiles. While calculated solid flux data quantitatively agreed more with experimental results at the riser center rather than at the wall, calculated granular temperature and solid viscosity trends agreed qualitatively with experiments. Gao et al. (2012) found a core-annulus flow structure from Eulerian-Eulerian simulations of a CFB riser using a steady-state multi-phase model. Predicted solid concentration and velocity profiles agreed well quantitatively with experimental results, and the inlet boundary conditions had negligible effect on the core-annulus flow. Also the solid motion was found to be dominated by individual particles in the core and by particle clusters in the annulus. Recently, Hou et al. (2016) simulated a CFB riser using a drag force based model and obtained the core-annulus structure. While, a general quantitative agreement was found between predicted and experimental radial voidage profiles, slight deviations from experimental values were obtained at the riser center.

Riser geometry is also found to exert a considerable effect on solids flow in a CFB riser. Regarding type of riser exit, it was found that constricted (Hirama et al., 1992) or abrupt exits like the blinded T-shaped exit (Hussain et al., 2013; Martin et al., 1992a; Martin et al., 1992b; Zhang et al., 2013) increased solid concentration at the riser top due to solids accumulation (Hussain et al., 2013; Martin et al., 1992b) which yielded a C-shaped concentration profile (Bai et al., 1992; Martin et al., 1992b). Gradual exits like the elbow-shaped exit (Harris et al., 2003) or a 90- degree exit with a 45-degree baffle (Bai et al., 1992; Hussain et al., 2013) were found to affect solid concentration at the top of the riser to a lesser extent. An L-shaped (90-degree) exit had an intermediate effect on solid concentration (Bai et al., 1992; Hussain et al., 2013; Zhang et al., 2013). Simulations (Shi et al., 2015; Van engelandt et al., 2011) agreed well with the above experimental observations. Regarding riser entry, Bai et al. (1992) found that a weak inlet restriction to the riser (fully opened valve) led to an increased solid concentration at the riser bottom and a strong inlet restriction (valve minimally opened) decreased solid

concentration at the riser bottom, while De Wilde et al. (2005) noted that a bottom gas inlet hindered gas-solid mixing and a side gas inlet aided it. Regarding riser diameter, D , it has been observed that with increasing D , the uniformity of axial solid concentration (Bai et al., 1992), the cross-sectional average solid concentration (Qi et al., 2008), length of the acceleration region (Monazam and Shadle, 2008) and the length of influence of an abrupt exit (Monazam et al., 2016; Pugsley et al., 1997) increase, while axial solid mixing decreases (Rhodes et al., 1991). Regarding riser height, H , it has been found that shorter risers may only possess the acceleration region (Xu and Gao, 2003) and the region of fully developed flow may not exist (Bai et al., 1992) for them. Further, for gradual exits, the section of riser influenced by the exit decreases with decreasing H (Bai et al., 1992).

Most of the literature describes the core-annulus structure as a radially segregated solids flow with a lean core consisting of upflowing particles and a thin dense annulus consisting of downflowing particles. However, in the case of dense phase flow, a core-annular solids distribution persists, but, instead of a downflow annulus, an upflow annulus was consistently observed (Bi and Zhou, 1993; Issangya, 1998; Issangya et al., 1997a; Issangya et al., 1997b; Karri and Knowlton, 1991; Schuurmans, 1980). Here, ‘dense phase flow’ refers to an overall solids flux, $G_s > 200 \text{ kg/m}^2\text{s}$ and a solids holdup of greater than 10%. A CFB operating under such conditions was defined by Bi and Zhou (1993) as a High Density Circulating Fluidized Bed (HDCFB). Bai et al. (1999) found that the statistical and chaotic analyses from the signals obtained from such HDCFB risers were markedly different from in the ones from more dilute CFB risers. Gupta and Berruti (1998) demarcated ‘dense phase flow’ and ‘fast fluidization’ by an empirical correlation developed from experimental data on Geldart Group A particles. Grace et al. (1999) subsequently proposed a new flow regime, using experimental data obtained mostly from Geldart Group A particles. They called it ‘Dense Suspension Upflow’ (DSU). The key feature of the DSU regime is an upflow annulus, while retaining the variation of radial

solids concentration as in a conventional core-annulus flow (i.e., dilute core and dense annulus). Issangya et al. (2000) further carried out measurements of local voidage in an HDCFB riser using Geldart Group A particles and found that, while voidage fluctuations were low at the core, they reached a peak value (i.e., maximum heterogeneity) at some distance away from the wall, which is in contrast with the conventional core-annulus flow whereby maximum heterogeneity was found at the wall. Subsequently, Kim et al. (2004) demarcated three different flow regimes for CFB risers, based on the flow direction of the annulus and the solids holdup across the riser cross-section at a given height, ϕ . They used previously published experimental data, most of which was from Geldart Group A particles. Many authors have investigated the ‘Dense Suspension Upflow’ (DSU) regime (Geng et al., 2013; Geng et al., 2014; Parssinen and Zhu, 2001; Wang et al., 2014; Wei et al., 1998), but with the focus largely on Geldart group A.

Fig. 8.1 shows a qualitative sketch of the radial solids volume fraction for various fluidization regimes in a CFB, described above. The letters ‘C’ and ‘A’ denote the core and annulus, respectively. The black, up-arrows denote upward solids movement and the red down-arrows denote downward solids movement. Three principal regimes can be identified: (i) Dense suspension upflow, which has the highest solids volume fraction and a dilute core and denser annulus, both exhibiting upflow, (ii) Fast fluidization, which has a moderate solids concentration with a dilute upward moving core and a denser downward moving annulus, and (iii) Pneumatic transport, which has the lowest, nearly uniform radial solids concentration with upflow throughout the cross section.

Fig. 8.1. Qualitative sketch of the radial solids volume fraction for three fluidization regimes, namely, High Density Circulating Fluidized Bed (HDCFB), Fast fluidization and Dilute pneumatic transport. ‘C’ denotes the core and ‘A’ denotes the annulus. The black, up-arrows denote solids upflow and the red down-arrows denote solids downflow.

Table 8.1 lists the important characteristics of the experimental systems used in some studies mentioned above. Although there has been some research on demarcating flow regimes in CFB risers containing Geldart group B particles (Das et al., 2008), none has focused on investigating the core-annulus behavior of these systems. Geldart Group B particles are well-acknowledged to fluidize differently from Geldart Group A particles, with the most significant

difference being the lack of cohesion in the former (Geldart, 1973). In particular, a previous study has shown surprisingly that Geldart Group B particles exhibited a reverse core-annulus behavior (i.e., a dense core and a dilute annulus) due to the higher Stokes numbers (St) of the particles (Chew et al., 2012b). Because behavioral trends of Geldart Group A particles cannot be directly extrapolated to predict those of Group B, an investigation into the hydrodynamics of the latter warranted especially with the increasing prominence in its use as inert materials to improve the fluidization quality of biomass (Cui and Grace, 2007; Oliveira et al., 2013; Paudel and Feng, 2013).

Table 8.1. Important characteristics of the experimental systems used in the studies mentioned in Section 8.2

Author	Riser diameter, D (m)	Riser height, H (m)	Particle species	Particle diameter, d_{ave} (μm)	Particle density, ρ_p (kg/m^3)	Geldart Group
Weinstein et al. (1986)	0.152	8.5	Engelhard cracking catalyst	20-120	1460	A
Monceaux et al. (1986)	0.144	8	FCC	59	N.A.	A
Dry (1986, 1987)	0.09	7.2	FCC	71	1370	A
Zhang et al. (1991)	0.032	2.8	FCC	54	929.5	A
	0.09	10	Hollow glass beads	75	607	A
	0.3	12	Alumina	42.8	2003	A
			Alumina	34.4	2003	A
Herb et al. (1992)	0.05	2.7	FCC	68	N.A.	A
	0.15	10.8	Sand	125	N.A.	N.A.
			Sand	276	N.A.	B

Author	Riser diameter, D (m)	Riser height, H (m)	Particle species	Particle diameter, d_{ave} (μm)	Particle density, ρ_p (kg/m^3)	Geldart Group
Rhodes and Laussmann (1992)	0.152	4	Alumina	74.9	2456	A
Bai et al. (1992)	0.186	8	FCC	54	1545	A
	0.14	8-12	Silica gel-A	280	706	A
			Silica gel-B	165	710	A
			Sand	31	2660	A
Yang et al. (1993)	0.14	11	FCC	54	1550	A
Brereton and Grace (1993)	0.152	9.3	Sand	148	2650	B
Zhou et al. (1994)	0.146 x 0.146	9.14	Sand	213	2640	B
Issangya et al. (1997a)	0.076 and 0.102	6.1 and 7.62	FCC	90	1600	A
Wei et al. (1998)	0.186	8	FCC	54	1398	A
Bai et al. (1999)	0.076 and 0.102	6.1 and 8.32	FCC	70	1600	A
Johnsson et al. (2000)	0.12 x 0.7	8.5	Sand	310	2600	B
Issangya et al. (2000)	0.076 and 0.102	6.1 and 9.14	FCC	70	1600	A
Parssinen and Zhu (2001)	0.076	10	FCC	67	1500	A
Kim et al. (2004)	0.203	5.9	FCC	70	1700	A
Geng et al. (2013); Geng et al. (2014)	0.1-0.2	10.06	FCC	80	1780	A

Author	Riser diameter, D (m)	Riser height, H (m)	Particle species	Particle diameter, d_{ave} (μm)	Particle density, ρ_p (kg/m^3)	Geldart Group
Wang et al. (2014)	0.076	10	FCC	76	1780	A

N.A. = Not available

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9

Analyzing differential pressure signals in a pilot-scale CFB riser

This chapter is targeted at assessing the hydrodynamic characteristics that can be understood from a comprehensive dataset of pressure signals via wavelet decomposition.

9.1 Experimental protocol

9.1.1 Experimental setup

Fig. 9.1 shows a schematic of the circulating fluidized bed located at PSRI's (Particulate Solid Research Incorporated) research facility in Chicago, USA, which was the same as that in previous efforts on characterizing clusters (Chew et al., 2012a, b), mass flux (Chew et al., 2011), species segregation (Chew and Hrenya, 2011), and radial concentration (Chew et al., 2012c). The CFB had a riser with an inner diameter of 0.30 m and was 18.3 m tall. The air required for fluidization was supplied by Atlas Copco blowers, and the air flow rate was measured with the differential pressure across an orifice plate, while the air temperature and relative humidity were measured using an Omega HX93AV-RP1 probe inserted flush with the wall before the mixing pot. The control of the particle flux in the riser was enabled by a pneumatic slide valve.

Twelve pressure taps, which were connected to 11 differential pressure (ΔP) transmitters, had snubbers (Omega.com) at the tips and were installed flush with the walls axially along the riser. To avoid possible entry of particles into the pressure taps, pressure snubbers (Omega.com) were placed at the tip of the pressure taps. Although snubbers are well-acknowledged to dampen the signals, their use is necessary to prevent the particles from entering and disrupting the pressure transmitters. It should be noted that the capability to obtain information from the dampened signal here indicates that getting similar information from the

more sensitive, undampened signals would be even more straightforward. All data, including pressure, temperature, relative humidity and superficial gas velocity (U_g), were recorded via a DASylab data acquisition program throughout the experiments.

Fig. 9.1. Schematic of the pilot-scale circulating fluidized bed (CFB) located at PSRI's research facility in Chicago, USA

9.1.2 Materials

Five Geldart Group B (Geldart, 1973) particle systems were investigated, namely, three monodisperse and two binary mixtures.

As listed in Table 9.1, the three monodisperse particles were chosen such that two had the same median diameter (d_{p50}) and two had the same particle density (ρ_p). The three monodisperse particle systems were small glass ($d_{p50} = 170 \mu\text{m}$, $\rho_p = 2500 \text{ kg/m}^3$), large glass ($d_{p50} = 650 \mu\text{m}$, $\rho_p = 2500 \text{ kg/m}^3$) and high-density polyethylene (HDPE; $d_{p50} = 650 \mu\text{m}$, $\rho_p = 900 \text{ kg/m}^3$). Glass beads of both sizes were procured from Midwest Finishing Systems and HDPE was procured from Dyneon LLC. Note that the term 'monodisperse' does not refer to

identically sized particles, which is difficult to obtain in large quantities, but to a diameter range that is as narrow as experimentally possible. To obtain a narrow particle size distribution (PSD), the particles procured were sieved using a Sweco industrial sieve. Representative samples obtained using a sample thief were further classified in a Ro-Tap sieve shaker to obtain characteristic PSD widths (i.e., ratio of the standard deviation (σ) of the mass-based PSD to d_{p50}). The σ/d_{p50} values of the ‘monodisperse’ small glass, large glass and HDPE were found to be 12%, 9% and 11%, respectively, as listed in Table 9.1.

The two binary mixtures were chosen such that one was a binary-size mixture (i.e., composed of two constituents varying in d_{p50} but not ρ_p) and the other was a binary-density mixture (i.e., composed of two constituents varying in ρ_p but not d_{p50}). As shown in Table 9.1, the binary-size mixture comprised of monodisperse small glass and monodisperse large glass at a 1:1 volume ratio, while the binary-density mixture comprised of monodisperse large glass and monodisperse HDPE at a 1:1 volume ratio.

Table 9.1. Properties of the five particle systems investigated

System	Material	Volume %	Median particle diameter, d_{p50} (μm)	σ/d_{p50} (%)	Particle density, ρ_p (kg/m^3)
Monodisperse	Small glass	100	170	12	2500
Monodisperse	Large glass	100	650	9	2500
Monodisperse	HDPE	100	650	11	900
Binary-size	Small glass	50	170	12	2500
	Large glass	50	650	9	2500
Binary-density	Large glass	50	650	9	2500
	HDPE	50	650	11	900

9.1.3 Operating conditions

The operating conditions investigated were the same as those in previous studies (Chew et al., 2011, 2012a, b, c; Chew and Hrenya, 2011), and chosen to be as wide a range as possible in view of the physical constraints of the fluidized material, experimental setup and instruments used. Two values of superficial gas velocities (U_g) and overall mass flux (G_s) (as an indication of the solids circulation rate) were considered, consequently yielding four operating conditions as given in Table 9.2. Solid loading (m), which is a dimensionless quantity and defined as the ratio of overall mass flux to gas flux, is also listed in Table 9.2. Note that no cognizable deviations on the operating conditions were reported.

Table 9.2. Operating conditions

Superficial gas velocity, U_g (m/s)	Mass flux, G_s (kg/m ² s)	Solid loading, m (-)
13.5	120	7.4
13.5	260	16.0
17.0	120	5.9
17.0	260	12.7

9.1.4 Measurement technique

Pressure signals from all 11 differential pressure (ΔP) transmitters along the riser were recorded via a DASYlab data acquisition program. For the small glass, HDPE and the binary-size and binary-density mixtures, data acquisition was at 1000 Hz and each data collection lasted 30 s. The measurement frequency was consistent with the values recommended in reviews by van Ommen et al. (2011) and Johnsson et al. (2000) who considered them adequate to capture the details of the fluidized bed. The problem of over-capture of noise was overcome partly through wavelet decomposition, which enabled focus on relevant frequencies. Even if

some noise filtered through, the objective of differentiating between material properties and operating conditions was satisfactorily achieved. The measurement duration of 30 s was found to be sufficient to capture the riser characteristics, as described in previous studies on the same apparatus, materials and operating conditions (Chew et al., 2012a, b, c). Also, it is consistent with the typical measurement durations used for wavelet decomposition (Bi, 2007; Ellis et al., 2003; Guenther and Breault, 2007; Ren et al., 2001; Tahmasebpour et al., 2013; van Ommen et al., 2011; Yang and Leu, 2009). Due to the sufficient time given for signal capture and the fully developed nature of the system when tested, the pressure sensor delay did not play a significant role in affecting results. It should be noted that, for large glass, which was initially tested in the CFB riser, data acquisition frequency was preliminarily set at 100 Hz, as opposed to the other particles (1000 Hz) and each data collection lasted 60 s to ensure sufficient datapoints for statistical analysis. However, as described in Section 9.2.2, this did not affect the analysis, since a decomposition scale of 1 was applied for the wavelet decomposition of large glass signals and that of 4 to analyze the other particles, both of which captured a similar range of frequencies (50-100 Hz and 62.5-125 Hz, respectively) and yielded identical energy profiles for an empty riser.

9.2 Analysis techniques

9.2.1 Discrete Fourier Transform (DFT)

Fourier analysis breaks down a signal into its constituent sinusoids in order of their frequencies. The Discrete Fourier Transform (DFT), $X(f)$, of a time series signal $x(t)$ consisting of N discrete points is given as

$$X(f) = \sum_{t=1}^N x(t)e^{-2\pi jtf} \quad (9.1)$$

where f is the signal frequency and j is the imaginary unit. Fast Fourier Transform is a common algorithm to obtain the Discrete Fourier Transform of a time series signal. The power spectrum of a signal, calculated by taking the square of $X(f)$, gives the contribution of each frequency to the total power of the signal. In this study, the Welch method (Welch, 1967) of estimating the power spectrum of a signal was used because of its advantage in reducing the variance (Tahmasebpour et al., 2013).

9.2.2 Wavelet decomposition

The raw pressure signals were analyzed using wavelet decomposition via the wavelet toolbox on MATLAB. At least 5 repeat datasets were analyzed for each particle system and operating condition. Wavelet decomposition was chosen because different frequency ranges of data signals can be extracted from the raw signal (Mallat, 1989, 2009). Specifically, wavelet decomposition breaks down a raw signal into ‘details’, the high frequency component, and ‘approximations’, the low frequency component. In the current work, Daubechies wavelets of order three (DB3) was used (Chew et al., 2012a). Fig. 9.2 illustrates the procedure of wavelet decomposition.

Fig. 9.2. Decomposition of signals via wavelet decomposition

Fig. 9.3 shows an example of the approximation and detail signals at various scales. As expected, the signal frequency decreased with each subsequent increase in scale (j), and the original timeline was retained throughout the decomposition. A thorough mathematical explanation of the wavelet decomposition technique can be found elsewhere (Mallat, 1989, 2009). Different scales of wavelet decomposition are known to be associated with different phenomena in the fluidized bed, with the highest, intermediate and lowest frequencies corresponding to noise, fluidization characteristics and systemic effects, respectively (Yang and Leu, 2009). The noise, fluidization characteristics and systemic effects are also termed micro-scale (i.e., high frequency or low j), meso-scale (i.e., intermediate frequency or intermediate j) and macro-scale (i.e., low frequency or high j), respectively (Ellis et al., 2003; Guenther and Breault, 2007; Li and Kwauk, 1994; Ren et al., 2001; Yang and Leu, 2009). Hence, the energy contained in a wavelet-decomposed detail signal at different scales of

decomposition can be used to quantify the significance of each of the components. The energy of a detail signal at decomposition scale j is given by (Chew et al., 2012a):

$$E_{Dj} = \sum_1^n [D_j(t)]^2 \quad (9.2)$$

where n is the total number of datapoints for raw signal collected, which is the same as that in the detail signal.

Fig. 9.3. The raw pressure signal was collected at 1000 Hz for 30 s for monodisperse HDPE at the operating condition of $U_g = 13.5$ m/s and $G_s = 120$ kg/m²s, and at $h/H = 0.389$. The approximations (A_j) and details (D_j) were obtained via wavelet decomposition at $j = 1, 4, 7$ and 11.

Fig. 9.4a shows a sample plot of the energies calculated using Eq. (9.2) of the wavelet-decomposed detail signals at increasing scales from 1 through 20. Specifically, the x -axis represents scales from 1 through 20, while the y -axis represents the respective energy of the details (E_{Dj}). The different axial positions at which raw pressure signals were obtained are

indicated by the legend, and each error bar represents the span of the energies from repeats. Fig. 9.4a indicates that the highest E_{Dj} values were from the intermediate scales (j) of 12 – 15, which is also termed the ‘meso-scale’ (Ellis et al., 2003; Guenther and Breault, 2007; Li and Kwauk, 1994; Ren et al., 2001; Yang and Leu, 2009) and contains information on fluidization characteristics like clusters (Chew et al., 2012a, b; Chew et al., 2013; Guenther and Breault, 2007). Unfortunately, the error bars at the meso-scale (i.e., $j = 12 - 15$) were large, hence drawing sound conclusions was problematic. Thereby, armed with the understanding that the pneumatic nature of pressure transmitters suggests that the air component at the lower scales (i.e., higher frequencies) (Wang et al., 2016) would provide significant information, scrutiny on the lower scales was carried out in Fig. 9.4b. Specifically, Fig. 9.4b zooms in only on $j = 1 - 5$ of Fig. 9.4a, and it is apparent that the error bars are very much reduced. Expectedly, E_{Dj} decreased with j steeply between $j = 1$ and 2 but started to plateau at $j = 4$, since the micro-scale is associated with noise, which becomes progressively less dominant as frequency decreases (i.e., j increases). For datasets collected at 1000 Hz, $j = 4$ had more reasonable error bars and presumably an adequate filtering out of noise inherent in the raw signal. Though the latter could not be verified with certainty, assuming reasonably that the noise level in the raw signal was constant, relative changes due to operating conditions and material properties would be observable. Thus, $j = 4$ was chosen as a suitable scale for further analyses. Note that the datasets of large glass were acquired at a frequency of 100 Hz preliminarily, while a higher frequency of 1000 Hz was subsequently used for acquiring those of monodisperse small glass, monodisperse HDPE and binary-size and binary-density mixtures (Chew et al., 2012c). Therefore, for a valid comparison among all five particle systems, the frequency ranges corresponding to the detail signal has to agree; because $j = 4$ means that D_4 for a raw data collected at 1000 Hz corresponds to a frequency range of 62.5 – 125 Hz (Fig. 9.2), the dataset collected at 100 Hz has to be analyzed at $j = 1$ since D_1 corresponds to a similar frequency

range of 50 – 100 Hz. Fig. 9.5 affirms this. Specifically, Fig. 9.5 shows two plots of dimensionless riser height (h/H) versus E_{Dj} for an empty riser at $U_g = 0$ and $G_s = 0$. One represents the E_{D4} trend for the data collected at 1000 Hz, while the other the E_{D1} trend for the data collected at 100 Hz. Each error bar represents the standard deviation of 25 repeats. Two important inferences from Fig. 9.5 are: (i) the magnitudes and shapes of both trends agree well, which validates a fair comparison between the usage of E_{D1} for large glass (whose data was collected at 100 Hz) and E_{D4} for the other materials (whose data was collected at 1000 Hz); and (ii) the rather peculiar non-monotonic relationship between E_{Dj} and h/H is an inherent characteristic of the riser and pressure transmitters, and not due to any variation in materials and/or operating conditions.

Fig. 9.4. E_{Dj} vs Scale (j) for the binary-density mixture at the operating condition of $U_g = 17$ m/s and $G_s = 260$ kg/m²s at various h/H : (a) Scales 1 through 20; and (b) Scales 1 through 5.

Fig. 9.5. h/H versus E_{Dj} when $U_g = 0$ and $G_s = 0$. E_{D1} for a data acquisition frequency of 100 Hz corresponds well with E_{D4} for a data acquisition frequency of 1000 Hz, thereby ensuring a fair comparison of the data collected at different frequencies. Each error bar represents the standard deviation of 25 replicates.

9.3 Average differential pressure

Fig. 9.6 displays dimensionless height (h/H , where h is the height evaluated and H is the total height of the riser) versus change in pressure with respect to change in height ($\Delta P/\Delta h$) for the five particle systems investigated (namely, three monodisperse and two binary systems). Each of the four sub-plots represents each operating condition, and each error bar represents the span of two repeat measurements. Four observations are worth highlighting. Firstly, the higher m conditions (Fig. 9.6b and d) led to higher maximum $\Delta P/\Delta h$ values at $h/H \approx 0$. Secondly, the noisiest profiles in terms of largest error bars were that of large HDPE presumably due to electrostatics and/or non-spherical shape factor. Thirdly, the impact of material property on $\Delta P/\Delta h$ was significant at $h/H < 0.2$ and $h/H < 0.4$ respectively for the lower (Fig. 9.6a and c) and higher (Fig. 9.6b and d) m conditions, and negligible at higher heights. Specifically, the demarcation of developing and fully developed flows in the riser appeared to be at $h/H \approx 0.2$

and $h/H \approx 0.4$ respectively for the lower (Fig. 9.6a and c) and higher (Fig. 9.6b and d) m conditions. Fourthly, an abrupt increase in the $\Delta P/\Delta h$ values at the top of the riser is only apparent for the large glass material at the higher U_g conditions (Fig. 9.6c and d), which indicates that exit effects are only prominent in these cases. The less dominant exit effects can be attributed to the rounded-elbow exit used in this setup, as opposed to a more abrupt one (Lim et al., 1995; Pugsley et al., 1997; Wu et al., 2010). This agrees with the ‘reverse’ core-annulus phenomenon reported earlier, whereby a denser core and more dilute annulus was observed for the larger materials at the riser top, which is reported as due to larger Stokes numbers coupled with exit effects (Chew et al., 2012c). As stated earlier, pressure transmitters are the most common instrumentation, and often the only, available for commercial fluidized beds, hence the capacity to extract information from pressure signals is crucial for both trouble-shooting and optimization purposes. The incapacity of a simple averaging as in Fig. 9.6 for obtaining more information needed for clearly discerning variations in the riser necessitates alternative analysis methods (Chew et al., 2015; Drahoš et al., 1988).

Fig. 9.6. Dimensionless height (h/H) versus $\Delta P/\Delta h$ for three monodisperse and two binary particle systems at four operating conditions: (a) $U_g = 13.5$ m/s, $G_s = 120$ kg/m²s, $m = 7.4$; (b) $U_g = 13.5$ m/s, $G_s = 260$ kg/m²s, $m = 16.0$; (c) $U_g = 17$ m/s, $G_s = 120$ kg/m²s, $m = 5.9$; (d) $U_g = 17$ m/s, $G_s = 260$ kg/m²s, $m = 12.7$.

9.4 Analysis via Discrete Fourier Transform

Fig. 9.7 shows the power spectra of the pressure signals of five particle systems, taken at four operating conditions (Table 9.2) and $h/H = 0.39$, as log-log plots of power versus frequency. As seen in Fig. 9.7, all materials at all the operating conditions exhibited the greatest power at frequencies below 0.1 Hz, and the power magnitudes decreased rapidly with increasing frequency. The dominant time period of longer than 10 s seems surprising, but three

plausible reasons may shed some light. The first relates to the significance of dominant frequency in CFBs. A number of authors have concluded that a dominant frequency cannot be clearly defined (Bai et al., 1996; Ishii and Horio, 1991; Johnsson et al., 2000; Luckos et al., 2007) for the transport conditions investigated here and little physical meaning (Johnsson et al., 2000; van Ommen et al., 2011) can be ascribed to it. Others have noted that CFBs at transport conditions lack temporal periodicity (Bai et al., 1999; Du et al., 2006; Luckos et al., 2007) and do not have a characteristic time scale (Yashen et al., 1992). The second reason is associated with the influence of solids near the wall of the riser. Many authors (Bouillard and Miller, 1994; Chalermssinsuwan et al., 2014; Drahoš et al., 1988; Ishii and Horio, 1991; Jaiboon et al., 2013; Zi et al., 2016; Zijerveld et al., 1998) have obtained the highest energy values in the power spectrums of CFBs at transport conditions at frequencies less than 0.1 Hz. This has been attributed to the increased presence of solids (Bouillard and Miller, 1994; Drahoš et al., 1988) or the movement of clusters (Jaiboon et al., 2013) at the riser wall. The third reason concerns the use of snubbers at the tip of the pressure taps. Snubbers are known to dampen signals, which may have contributed to the longer time periods obtained here. However, the power spectra were deficient in distinguishing the change in material or operating condition. It should also be noted that the power spectra of all the materials at all operating conditions were also similar at all the axial positions (h/H) investigated. Discrete Fourier Transform has been extensively employed to detect regime changes in fluidization (Drahoš et al., 1988; He et al., 2014; Jaiboon et al., 2013; Johnsson et al., 2000; Satija and Fan, 1985; Tahmasebpour et al., 2013; van der Schaaf et al., 1999), but such changes are relatively more drastic compared to the differences in material property or operating conditions, which are more likely encountered in practical applications. Hence, both the simple averaging of the pressure signals (Fig. 9.6) and Discrete Fourier Transform (Fig. 9.7) appear to be inadequate in differentiating more subtle changes in the riser like operating condition and material property.

Fig. 9.7. Power spectra obtained by Discrete Fourier Transform of the differential pressure (ΔP) signals for three monodisperse and two binary particle systems at $h/H = 0.39$ at four operating conditions: (a) $U_g = 13.5$ m/s, $G_s = 120$ kg/m²s, $m = 7.4$; (b) $U_g = 13.5$ m/s, $G_s = 260$ kg/m²s, $m = 16.0$; (c) $U_g = 17$ m/s, $G_s = 120$ kg/m²s, $m = 5.9$; and (d) $U_g = 17$ m/s, $G_s = 260$ kg/m²s, $m = 12.7$.

9.5 Analysis via wavelet decomposition

In view of the apparent deficiency of the raw $\Delta P/\Delta h$ values and Discrete Fourier Transform in differentiating operating conditions and material properties, wavelet decomposition was harnessed to further probe the pressure signals. The impact of operating

condition, material property and axial position was analyzed via wavelet decomposition of the ΔP signals. In each subplot in the following, the x -axis reflects the energy of each scale of the detail signal (E_{Dj} ; calculated using Eq. (9.2)), while the y -axis represents the dimensionless riser height (h/H , where h is the riser height evaluated and H is the total riser height).

9.5.1 Impact of operating condition

Fig. 9.8 presents h/H versus E_{Dj} for the five particle systems at four operating conditions. Note that $j = 1$ for the monodisperse large glass and $j = 4$ for all the other materials, because the data acquisition frequency was an order-of-magnitude lower (i.e., 100 Hz) for the monodisperse large glass. As seen from Fig. 9.5, the peculiar non-monotonic E_{Dj} trends in the h/H versus E_{Dj} plots of Fig. 9.8 were characteristic of the pressure transmitters and riser. Nonetheless, Fig. 9.8 clearly shows that the E_{Dj} trends were distinctly different for the different operating conditions throughout the riser, which agree with that of $\Delta P/\Delta h$ in Fig. 9.6 at the lower h/H values. Recalling earlier that the delineation of the developing and fully-developed flow in the riser was observed at about $h/H \approx 0.3$ (Fig. 9.6), it appears that the change in the flow regime axially along the riser was not apparent in the E_{Dj} profiles in Fig. 9.8.

Fig. 9.8. h/H versus E_{Dj} at four operating conditions for three monodisperse and two binary particle systems: (a) small glass; (b) large glass; (c) HDPE; (d) binary-size mixture; and (e) binary-density mixture. Note that $j = 1$ for the monodisperse large glass (b) and $j = 4$ for all the other materials (a, c, d and e), because the data acquisition frequency was an order-of-magnitude lower (i.e., 100 Hz) for the monodisperse large glass (b).

Specifically on Fig. 9.8, five observations are noteworthy: (i) the lower solids to gas flux ratios (m) tended to give higher E_{Dj} values, except for HDPE (Fig. 9.8c) which exhibited greater variations as represented by the larger error bars, which indicate that a lower m value is associated with a higher proportion of gas-phase information in the pressure signal; (ii) the large glass system (Fig. 9.8b) exhibited the most distinctly different E_{Dj} values between the lower and higher G_s conditions, which implied that the pressure signals of the larger and denser particles tended to be most sensitive to operating conditions; (iii) the largest error bars are found in Fig. 9.8c for HDPE due presumably to electrostatics and/or non-spherical shape effects, thereby the trends of E_{Dj} with respect to m did not agree with the other materials; (iv) the influence of operating conditions on E_{Dj} values tended to be apparent throughout the riser (Fig. 9.8), rather than only at the riser bottom for the $\Delta P/\Delta h$ values in Fig. 9.6; and (v) the small glass (Fig. 9.8a) and large glass (Fig. 9.8b) systems gave respectively the lowest and highest E_{Dj} values.

9.5.2 Impact of material property

Fig. 9.9 displays h/H with respect to E_{Dj} to showcase the impact of material property on the E_{Dj} values extracted from the pressure signals. Four interesting observations include: (i) the variations of E_{Dj} values due to the different materials were greater for the lower solids to gas flux ratio (m) conditions (Fig. 9.9a and c), which is in sync with the earlier observation that lower m values are tied with higher proportions of gas-phase behavior in the pressure signal; (ii) similar to that of the impact of operating condition (Fig. 9.6), material property tended to affect the E_{Dj} values to approximately the same extents throughout the riser; (iii) as noted earlier, at all operating conditions (Fig. 9.9a – d), the highest E_{Dj} values were for large glass followed by the binary-density system, while the lowest were for small glass, which implies

that the pressure signals from the monodisperse large glass system contained the most significant proportion of gas-phase information; and (iv) while the E_{Dj} profile of the binary-density system tended to mimic large glass (i.e., the denser constituent), the E_{Dj} profile of the binary-size system tended to mimic small glass (i.e., the smaller constituent).

Fig. 9.9. h/H versus E_{Dj} for three monodisperse and two binary particle systems at four operating conditions: (a) $U_g = 13.5$ m/s, $G_s = 120$ kg/m²s, $m = 7.4$; (b) $U_g = 13.5$ m/s, $G_s = 260$ kg/m²s, $m = 16.0$; (c) $U_g = 17$ m/s, $G_s = 120$ kg/m²s, $m = 5.9$; (d) $U_g = 17$ m/s, $G_s = 260$ kg/m²s, $m = 12.7$. Note that $j = 1$ for the monodisperse large glass and $j = 4$ for all the other materials, because the data acquisition frequency was an order-of-magnitude lower (i.e., 100 Hz) for the monodisperse large glass.

Regarding (iii), this can be related to Stokes number (St), which is the ratio of the inertia of the solid particle to the viscous forces of the gas, and has been used previously as an explanation for the reverse core-annulus profile (i.e., a denser core and a more dilute annulus) observed for all conditions for the large glass (i.e., highest St), some conditions for the HDPE (i.e., intermediate St) but not for the small glass (i.e., lowest St) (Chew et al., 2012c). Because particles with larger St values have a greater response time to changes in the gas streamlines, they tend to follow fluid streamlines less closely (McKeon et al., 2007; Yamamoto et al., 2001). With respect to the pressure signals, which is based on pneumatic disturbances, the lesser adherence of the particles with higher St values (e.g., large glass) to the gas flow implies that the pressure signals capture less of the particle contribution, hence the gas-phase contribution increases. Also, the relationship between the E_{Dj} values and particle diameter is tied to gas turbulence, as simulations have shown that larger particles enhance turbulence in the gas phase (Bolio et al., 1995; Rao et al., 2012), which hence increases E_{Dj} .

As for (iv), the binary-density mixture exhibited higher E_{Dj} values than binary-size, since the former had a higher characteristic St than the latter, which agree with the earlier discussion on (iii). It is compelling to compare with other behaviors of the binary mixtures characterized of the same materials, operating conditions and riser: (a) for mass flux profiles, the binary-density mixture mimicked the monodisperse large glass, whereas the binary-size mixture mimicked neither of its constituents (Chew et al., 2011); (b) for cluster duration, the binary-density mixture mimicked neither of its constituents, while the binary-size mixture mimicked monodisperse large glass (Chew et al., 2012b); and (iii) for cluster frequency, the binary-density mixture mimicked monodisperse HDPE, while the binary-size mixture mimicked monodisperse small glass (Chew et al., 2012b). Thereby, the collective results indicate that the binary-density mixture mimicked large glass for both mass flux and E_{Dj} values

of pressure signals, whereas the binary-size mixture mimicked small glass for cluster frequency and E_{Dj} values of pressure signals.

9.5.3 Impact of axial position

In order to understand the effect of axial position, the deviation of the E_{Dj} profile of a particular material from that of the empty column was determined. Fig. 9.10 displays h/H versus $E_{Dj} - E_{Dj,empty}$ (whereby $E_{Dj,empty}$ represents E_{Dj} when $U_s = 0$ and $G_s = 0$) for the five materials in five subplots at four operating conditions. The more vertical the $E_{Dj} - E_{Dj,empty}$ profile is, the less significant the impact of axial position is. Fig. 9.10 shows that all the $E_{Dj} - E_{Dj,empty}$ profiles are approximately vertical, which implies that the impact of axial position on the E_{Dj} profiles was secondary to that of operating conditions (Fig. 9.8) and material properties (Fig. 9.9). Note that previous efforts on characterizing the same materials under the same operating conditions in the same riser indicated in contrast that axial position had the greatest influence (relative to operating condition and material property) on the shape of profiles of cluster characteristics (namely, cluster appearance probability, duration and frequency) (Chew et al., 2012a, b) and mass flux (Chew et al., 2011).

Fig. 9.10. h/H versus $E_{Dj} - E_{Dj,empty}$ at four operating conditions for three monodisperse and two binary particle systems: (a) small glass; (b) large glass; (c) HDPE; (d) binary-size mixture; and (e) binary-density mixture. Note that $j = 1$ for the monodisperse large glass (b) and $j = 4$ for all the other materials (a, c, d and e), because the data acquisition frequency was an order-of-magnitude lower (i.e., 100 Hz) for the monodisperse large glass (b).

9.6 Conclusion

Differential pressure (ΔP) signals were recorded at 11 axial positions along a pilot-scale CFB riser for five different particle systems (namely, three monodisperse materials and two binary mixtures) and four operating conditions. The effects of material property, operating condition and riser axial position on the ΔP signals were evaluated.

A simple averaging of the ΔP signals axially along the riser led to the $\Delta P/\Delta h$ profiles, which only showed variations due to operating conditions and particle properties lower in the riser. Power spectra of the ΔP signals obtained via Discrete Fourier Transform too were not sensitive enough to differentiate between varying material properties and operating conditions. Hence, wavelet decomposition was harnessed to further probe the ΔP signals. In part because pressure transmitters senses pneumatic disturbances, the higher-frequency (lower-scale) micro-scale associated with air rather than the lower-frequency (higher-scale) meso-scale associated with particles was used in this effort. Thereby, the energy contained in the decomposed signals (E_{Dj}) was employed to provide indications of fluidization under various conditions. In contrast to the $\Delta P/\Delta h$ values, which were insensitive to operating conditions or material property higher in the riser, the influence of operating conditions and material on E_{Dj} values tended to be apparent throughout the riser.

The E_{Dj} trends show that: (i) higher E_{Dj} values were obtained at lower solids to gas flux ratio (m) conditions, which implies reasonably that the gas-phase contribution was higher for the more dilute operating conditions; (ii) E_{Dj} values were more sensitive to material properties at the lower m conditions; (iii) among monodisperse particles, the E_{Dj} values of larger and denser particles (i.e., large glass in this case) tended to be most sensitive to operating conditions, which can be traced to Stokes number and gas turbulence effects; (iv) among the five particle

systems, monodisperse small glass and monodisperse large glass systems gave respectively the lowest and highest E_{Dj} values, which indicate the least significant gas-phase contribution (and hence the most significant particle-phase contribution) to the ΔP signals for the former; (v) while the E_{Dj} profile of the binary-density system tended to mimic the denser constituent (i.e., large glass), the E_{Dj} profile of the binary-size system tended to mimic the smaller constituent (i.e., small glass), which implies that particle density exerts a more dominant influence than particle diameter on ΔP signals; and (vi) the effect of axial position on E_{Dj} was secondary compared to that of operating condition and material property.

Collectively, these observations provide additional insights not only into the mechanistic relationship between the gas phase and the particles, but also information available from the ubiquitous pressure transmitters in fluidized bed systems which can find potential use in practical operations. Regarding the former, the enhanced turbulence caused by larger particles have been observed in simulations (Bolio et al., 1995; Rao et al., 2012), but has been difficult to experimentally investigate. The higher E_{Dj} values of the large glass beads (i.e., the larger, denser monodisperse system) revealed in this work provides experimental evidence of the phenomenon. As for the latter, since the influence of operating conditions and material properties on E_{Dj} values was found to be detectable throughout the riser, unlike the $\Delta P/\Delta h$ values, a change in the hydrodynamics at any height in the riser can be readily monitored through E_{Dj} values, which is beneficial for trouble-shooting, optimization and process control.

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10

Annulus flow behavior of Geldart Group B particles in a pilot-scale CFB riser

This chapter studies the annulus flow behavior of Geldart Group B particles in a CFB riser with respect to the effects of material property, operating conditions and polydispersity. A comparison against available regime maps is also done.

10.1 Experimental protocol

The CFB riser used is identical to the one in the previous study. The reader is referred to Section 9.1.1 for details.

10.1.1 Materials

Six particle systems, all belonging to Geldart Group B, were investigated: three monodisperse systems, with two among them having different particle diameters (d_{ave}) and two among them having different particle densities (ρ_p); two binary mixtures, one consisting of constituents differing in d_{ave} (i.e., binary-size) and one consisting of constituents differing in ρ_p (i.e., binary-density); and a continuous particle size distribution (PSD). All particles were approximately spherical. Table 10.1 lists the particle systems investigated and the relevant properties.

Table 10.1. Properties of the materials investigated

System Type	PSD Width, σ/d_{ave} (%)	Material	Volume %	Average particle diameter, d_{ave} (μm)	Particle density, ρ_p (kg/m^3)
Monodisperse	12	Small glass	100	170	2500
Monodisperse	9	Large glass	100	650	2500
Monodisperse	11	HDPE	100	650	900
Binary-size	12	Small glass	50	170	2500
	9	Large glass	50	650	2500
Binary-density	9	Large glass	50	650	2500
	11	HDPE	50	650	900
Continuous PSD	25	Small glass	100	170	2500

In the case of monodisperse materials, small glass ($\rho_p = 2500 \text{ kg/m}^3$ and $d_{ave} = 170 \mu\text{m}$), large glass ($\rho_p = 2500 \text{ kg/m}^3$ and $d_{ave} = 650 \mu\text{m}$) and high density polyethylene, HDPE ($\rho_p = 900 \text{ kg/m}^3$ and $d_{ave} = 650 \mu\text{m}$), were investigated. While small glass and large glass had the same ρ_p but different d_{ave} , large glass and HDPE had the same d_{ave} but different ρ_p . As it was not experimentally possible to obtain identically sized particles, efforts were made to obtain as narrow a size distribution as possible. Glass beads of both sizes from Midwest Finishing Systems and HDPE from Dyneon LLC were sieved using a Sweco industrial sieve. The PSD widths, defined as the ratio of the standard deviation (σ) of mass-based PSD to d_{ave} , of monodisperse small glass, large glass and HDPE were 12%, 9% and 11%, respectively, as listed in Table 10.1. Binary mixtures were prepared by mixing two monodisperse materials in a one-to-one volume ratio, while the continuous PSD ($\sigma/d_{ave} = 25 \%$) was used as obtained from the vendor.

10.1.2 Operating conditions

The operating conditions investigated were selected with considerations of the test unit constraints, as detailed elsewhere (Chew et al., 2011a; Chew et al., 2012c). Tests for all the six particle systems were conducted at two superficial gas velocities (U_g) and two mass fluxes (G_s) in the riser as given in Table 10.2. The superficial gas velocity (U_g) values were determined by an orifice plate (as mentioned in Section 9.1.1), while the overall mass flux (G_s) was calculated by summing up the local mass flux measurements via an extraction probe (as will be detailed in Section 10.1.3). The inlet temperature of the gas was in the range of 15–25 °C, while the pressure profiles have been reported in a previous study (Chew et al., 2012c). The solid loading ratio (m), a dimensionless quantity defined as the ratio of overall mass flux to gas mass flux, is also given in Table 10.2. Note that no cognizable deviations on the operating conditions were reported.

Table 10.2. Operating conditions for the different materials

Material	Superficial gas velocity, U_g (m/s)	Mass flux, G_s (kg/m ² s)	Solids loading ratio, m (-)
Monodisperse materials and binary mixtures	13.5	120	7.4
	13.5	260	16.0
	17.0	120	5.9
	17.0	260	12.7
Continuous PSD	10.0	50	4.2
	10.0	300	25.0
	15.0	50	2.8
	15.0	300	16.7

10.1.3 Mass flux measurement

The method of measuring the local mass flux in the riser has been detailed elsewhere (Chew et al., 2011b). In brief, the upward and downward local mass flux values ($G_{r,upward}$ and $G_{r,downward}$, respectively) at a given radial location were measured using an extraction probe. The extraction probe was operated non-isokinetically (Aguillón et al., 1995; Chew et al., 2011b; Rhodes and Laussmann, 1992; Salvaterra et al., 2005; Zhang et al., 1997), and it was verified that suction velocity did not affect the results, which was expected of Group B particles as they do not follow gas streamlines closely due to relatively higher Stokes numbers associated with them (Chew et al., 2011b, 2012c). The global error of the probe was determined to be within an acceptable tolerance of $\pm 10\%$. Specifically, two checks were applied: (i) consistency of measurements over repeated runs and varying time durations and (ii) consistency of net G_s across the cross section across the riser height; this ensures a mass balance across the riser. Regarding (i) it was found after multiple runs at various time durations that at durations greater than 20 s, the net G_s measurements varied within an acceptable tolerance of $\pm 10\%$ regardless of measurement duration. Regarding (ii), a target G_s which was the same as the operating conditions (e.g. 260 kg/m²s), which could be controlled via a pneumatic slide valve in the bottom portion of the re-circulating loop, was set. Due to conservation of mass, the net G_s measurements should be consistent across the riser height with the value of the target (i.e. 260 kg/m²s), which they were, within a reasonable $\pm 10\%$ tolerance. This ensured that the mass flux measurements were correct. Interested readers are referred elsewhere (Chew et al., 2011a) for details. To allow for easier comparison across the different G_s conditions, the local mass flux was normalized with respect to the overall mass flux (G_s), which is expressed as follows (Chew et al., 2011b):

$$G_{r,net,norm} = \frac{G_{r,upward} - G_{r,downward}}{G_s} \quad (10.1)$$

whereby G_s is the summation of the net flux measured at 11 radial positions across the riser, as detailed in a previous study (Chew et al., 2011b). This normalization also helped minimize errors, hence the error bars in Figs. 10.1-10.5 are too small to be discernable.

Because this study only focuses on annulus flow behavior, only the $G_{r,net,norm}$ values at $r/R = 0.94$ (where r is the radial position assessed and R is the radius of the riser), which is the radial position closest to the wall where the measurement was taken, were taken into account.

10.2 Annulus mass flux profiles

In view of the focus of this study on understanding annulus flow behavior, only the mass flux near the wall was investigated. Specifically, this analysis was targeted at identifying the operating conditions, riser axial positions and particle properties (i.e., material type, polydispersity type) at which the particle systems examined exhibited upflow (i.e., positive $G_{r,net,norm}$) or downflow (i.e., negative $G_{r,net,norm}$) annulus behavior. The development of a regime map was beyond the scope of this study, but instead the current data was juxtaposed onto the available regime maps to gauge the applicability or lack thereof to Geldart Group B particles.

The effect of the particle properties of monodisperse small and large glass, and HDPE particles on the annulus is showed in Fig. 10.1. Normalized riser axial position, h/H (where h is the height evaluated and H is the total height of the riser), is plotted against the normalized radial solids flux, $G_{r,net,norm}$, at $r/R = 0.94$. The properties of the large glass, small glass and HDPE particles are given in Table 10.1. Each sub-plot represents a given operating condition (Table 10.2). Each error bar represents the span of values obtained from at least two repeat measurements, although the error bars are largely too small due to the normalization to be

clearly noticeable. A negative $G_{r,net,norm}$ value means downflowing of solids in the annulus, while a positive $G_{r,net,norm}$ value means upflowing of solids in the annulus. Five observations from Fig. 10.1 are worth highlighting. Firstly, regarding the solids flow direction in the annulus, the larger glass and HDPE particles exhibited an upflow annulus at all operating conditions, whereas the smaller glass particles gave both an upflow and a downflow annulus behavior. This suggests that particle diameter (d_{ave}) rather than particle density (ρ_p) played a more dominant role in dictating upflow annulus behavior. Secondly, regarding the effect of particle density (ρ_p), the $G_{r,net,norm}$ values of the large glass and HDPE were largely similar, except at $h/H = 0.16$ for the higher G_s conditions (Fig. 10.1b and d), which implies that the variations in annulus behavior due to particle density were minimal higher up in the riser. Thirdly, regarding the effect of particle diameter, the $G_{r,net,norm}$ values of small glass and large glass were similar higher up in the riser ($h/H \geq 0.47$) at lower G_s conditions (Fig. 10.1a and c), and similar at the lowermost ($h/H = 0.16$) and uppermost ($h/H = 0.92$) heights of the riser at the higher G_s conditions (Fig. 10.1b and d). This reflects that the impact of particle diameter on annulus flow behavior was significant lower in the riser at lower G_s conditions and middle of the riser at higher G_s conditions. Fourthly, the $G_{r,net,norm}$ values of the three particle types were consistently similar across the operating conditions tested only at the highest h/H of 0.92, which presumably indicates the dominance of exit effects (Chew et al., 2012c; De Wilde et al., 2003; Gupta and Berruti, 2000; Pugsley et al., 1997; Van der Meer et al., 2000) relative to particle property or operating condition. Fifthly, regarding axial variation of the annulus flow, the acceleration effects lower in the riser are clear, as indicated by the greater axial variation lower in the riser and the relatively invariant profiles above mid-height of the riser. In particular, the small glass exhibited the greatest axial variation at the lower G_s conditions, which suggests that the annulus flow of the small glass was more affected by the acceleration effects at these conditions than the larger particles. It should be noted that the acceleration-deceleration and/or

entrance-exit effects along the riser were more influential on the mass flux (Chew et al., 2011b) and cluster (Chew et al., 2012a, b) trends characterized for the same riser, materials and operating conditions, where the impact of axial position was the most dominant (relative to material properties and/or operating conditions).

Fig. 10.1. Dimensionless height (h/H) versus normalized radial solids flux ($G_{r,net,norm}$) at $r/R = 0.94$ for three monodisperse particle types at four operating conditions: (a) $U_g = 13.5$ m/s, $G_s = 120$ kg/m²s, $m = 7.4$; (b) $U_g = 13.5$ m/s, $G_s = 260$ kg/m²s, $m = 16.0$; (c) $U_g = 17$ m/s, $G_s = 120$ kg/m²s, $m = 5.9$; and (d) $U_g = 17$ m/s, $G_s = 260$ kg/m²s, $m = 12.7$. Note that negative and positive $G_{r,net,norm}$ values represent respectively downflow and upflow annulus.

Analogous to Fig. 10.1, Fig. 10.2 plots h/H versus $G_{r,net,norm}$ values at $r/R = 0.94$ for the binary-size and binary-density particle mixtures (Table 10.1). The common constituent in these mixtures was the large glass particle constituent. Four observations can be made. Firstly, both binary mixtures exhibited an upflow annulus, except for the binary-size mixture at $h/H = 0.27$

at the lower U_g conditions (Fig. 10.2a and b). This indicates that the larger constituent in the binary-size mixture dominated in dictating the upflow annulus behavior. Secondly, the $G_{r,net,norm}$ values for the two mixtures were similar throughout the riser at the lowest solid loading condition (Fig. 10.2c), indicating minimal impact of particle properties at this condition, which is consistent with Fig. 10.1c. Thirdly, the two mixtures each exhibited similar $G_{r,net,norm}$ trends with respect to h/H under the three higher solid loading conditions (Fig. 10.2a, b and d), which implies a negligible impact of operating conditions. Fourthly, the difference in $G_{r,net,norm}$ values between the mixtures were minimal higher up in the riser ($h/H \geq 0.73$) at all operating conditions, which indicate the dominating influence of riser position over particle property or operating condition. This is similar to that in Fig. 10.1 except that it was restricted only to $h/H = 0.92$. A note that the larger error bars at $h/H = 0.73$ for the lowest solid loading (Fig. 10.2c) relative to other measurements can be attributed to random fluctuations in local voidage, a characteristic of HDCFBs, which has been observed previously (Bai et al., 1999; Grace et al., 1999).

Fig. 10.2. Dimensionless height (h/H) versus normalized radial solids flux ($G_{r,net,norm}$) at $r/R = 0.94$ for the two binary mixtures at four operating conditions: (a) $U_g = 13.5$ m/s, $G_s = 120$ kg/m²s, $m = 7.4$; (b) $U_g = 13.5$ m/s, $G_s = 260$ kg/m²s, $m = 16.0$; (c) $U_g = 17$ m/s, $G_s = 120$ kg/m²s, $m = 5.9$; and (d) $U_g = 17$ m/s, $G_s = 260$ kg/m²s, $m = 12.7$.

Fig. 10.3 compares the $G_{r,net,norm}$ profiles at $r/R = 0.94$ of the binary-size mixture with its constituent small glass and large glass particles while Fig. 10.4 compares that of the binary-density mixture with its constituent particle species, large glass and HDPE. It can be seen in Fig. 10.3 that (i) despite the monodisperse small glass particles giving both upflow and downflow annulus, the binary-size mixture appeared to have a largely upflow annulus, which suggests a more dominant influence of the large glass constituent in dictating the upflow annulus behavior; (ii) other than the lowest solid loading condition (Fig. 10.3c) which was found to exhibit minimal influence of particle type (Figs. 10.1c and 10.2c), the binary-size mixture largely showed a different $G_{r,net,norm}$ trend from that of either constituent; and (iii) the $G_{r,net,norm}$ values of the binary-size mixture at $h/H = 0.16$ was distinctly greater than that of either constituent,

which indicates that the interaction between two species of different sizes in the binary mixture significantly increased the $G_{r,net,norm}$ value. It is interesting to note from (i) and (ii) that, though the large glass particles played a dominant role in dictating the upward direction of the annulus solids of the binary-size mixture, the $G_{r,net,norm}$ profiles of the binary-size mixture did not mimic those of the large glass constituent. For the binary-density mixture, Fig. 10.4 shows that (i) the $G_{r,net,norm}$ values of all three particle systems agreed under all four operating conditions higher in the riser ($h/H \geq 0.16$), which indicates that the binary-density mixture behaved like its constituents; and (ii) at the lowermost h/H position of 0.16, the binary-density mixture gave $G_{r,net,norm}$ values in between the two constituents at the higher G_s conditions (Fig. 10.4b and d). $G_{r,net,norm}$ values were either lower or higher than those of the two constituents at the lower G_s conditions (Fig. 10.4a and c). Juxtaposing Figs. 10.3 and 10.4 reveal that the binary-density mixture mimicked both its constituent particle species more than that of the binary-size, suggesting that the wall had a more significant effect on the interactions between species of different particle diameters rather than particle densities. This agrees with previously reported results (Chew et al., 2011b) where the radial mass flux profiles of the binary-size mixture did not mimic either constituent, but that of the binary-density mixture tended to mimic the large glass constituent.

Fig. 10.3. Dimensionless height (h/H) versus normalized radial solids flux ($G_{r,net,norm}$) at $r/R = 0.94$ for the binary-size mixture and its constituents (namely, small glass and large glass) at four operating conditions: (a) $U_g = 13.5$ m/s, $G_s = 120$ kg/m²s, $m = 7.4$; (b) $U_g = 13.5$ m/s, $G_s = 260$ kg/m²s, $m = 16.0$; (c) $U_g = 17$ m/s, $G_s = 120$ kg/m²s, $m = 5.9$; and (d) $U_g = 17$ m/s, $G_s = 260$ kg/m²s, $m = 12.7$.

Fig. 10.4. Dimensionless height (h/H) versus normalized radial solids flux ($G_{r,net,norm}$) at $r/R = 0.94$ for the binary-density mixture and its constituents (namely, large glass and HDPE) at four operating conditions: (a) $U_g = 13.5$ m/s, $G_s = 120$ kg/m²s, $m = 7.4$; (b) $U_g = 13.5$ m/s, $G_s = 260$ kg/m²s, $m = 16.0$; (c) $U_g = 17$ m/s, $G_s = 120$ kg/m²s, $m = 5.9$; and (d) $U_g = 17$ m/s, $G_s = 260$ kg/m²s, $m = 12.7$.

Fig. 10.5 illustrates h/H versus $G_{r,net,norm}$ at $r/R = 0.94$ for the continuous particle size distribution (PSD) of the small glass particles at $U_g = 10$ and 15 m/s and $G_s = 50$ and 300 kg/m²s. The four tests had solid loadings of 2.8 , 4.2 , 16.7 and 25.0 . Comparing the lower G_s conditions (Fig. 10.5a and c), it can be seen that the tendency for an upflow annulus was greater at higher U_g (Fig. 10.5c). Similarly, for the higher G_s conditions (Fig. 10.5b and d) too, the tendency for an upflow annulus increased with increasing U_g , though to a relatively lesser extent than that at the lower G_s conditions. This suggests that, for continuous PSDs, superficial gas velocity (U_g) was the more dominant factor than overall solids mass flux in dictating upflow annulus behavior.

Fig. 10.5. Dimensionless height (h/H) versus normalized radial solids flux ($G_{r,net,norm}$) at $r/R = 0.94$ for the continuous PSD of small glass at four operating conditions: (a) $U_g = 10$ m/s, $G_s = 50$ kg/m²s, $m = 4.2$; (b) $U_g = 10$ m/s, $G_s = 300$ kg/m²s, $m = 25.0$; (c) $U_g = 15$ m/s, $G_s = 50$ kg/m²s, $m = 2.8$; and (d) $U_g = 15$ m/s, $G_s = 300$ kg/m²s, $m = 16.7$.

It is reasonable to infer the impact of the continuous PSD vis-à-vis the monodisperse small glass systems by comparing (i) Fig. 10.1b ($m = 16$) with Fig. 10.5d ($m = 16.7$), and (ii) Fig. 10.1c ($m = 5.9$) with Fig. 10.5a ($m = 4.2$), each pair of which is at nearly similar solid loadings. Regarding (i), the $G_{r,net,norm}$ trends were very similar both in terms of magnitude and variation with respect to h/H , which indicates the presence of a continuous PSD did not change the annulus flow behavior at $m \approx 16$. However, at the lower m condition assessed in (ii), the monodisperse small glass exhibited distinct upflow annulus behavior at $h/H \geq 0.27$ (Fig. 10.1c), whereas the continuous PSD exhibited a clear upflow annulus behavior only at $h/H = 0.92$ (Fig. 10.5a), which suggests that the impact of continuous PSD was significant at lower solid loading (m) conditions. More specifically for the continuous PSD of small glass in Fig. 10.5, the highest

solid loading ($m = 25.0$) condition (Fig. 10.5b) gave the most marked downflow annulus behavior in terms of the most negative $G_{r,net,norm}$ value at $h/H = 0.47$, while the lowest solid loading ($m = 2.8$) condition (Fig. 10.5c) exhibited upflow annulus throughout the riser. It is worth pointing out that the radial mass flux profile for the continuous PSD of small glass was observed to be distinctly different at the lowest m condition relative to the other three conditions, which was different from the trends for the monodisperse small glass system, and was attributed either to behavioral differences due to polydispersity or the wider range of operating conditions (Chew et al., 2011b).

10.3 Regime maps

A regime map of CFB riser flow to demarcate conditions whereby core-annulus flow prevails was developed by Bi and Grace (1995). Three principal regimes were identified in order of decreasing superficial gas velocity (U_g), namely, homogeneous dilute-phase flow, core-annulus flow and fast fluidization. The homogeneous dilute-phase flow regime was described as having minimal lateral variation of solid concentration, with uniform upflow across the entire cross-section. The core-annulus flow regime, was delineated as having a lean upflow core and a denser downflow annulus. The fast fluidization regime was described as having a significant axial variation of solid concentration, with a dense, turbulent bottom and a core-annulus top. The demarcation between the homogeneous dilute-phase flow and core-annulus regimes, the minimum pressure velocity, V_{mp} , was defined (Bi and Fan, 1991) as;

$$V_{mp} = 10.1(gd_p)^{0.347} \left(\frac{G_s}{\rho_g}\right)^{0.31} \left(\frac{d_p}{D}\right)^{-0.139} Ar^{-0.021} \quad (10.2)$$

where G_s is the overall solids circulation mass flux in the riser, ρ_g is the gas density (1.2 kg/m^3), d_p is the particle diameter. Note that in Eqs. (10.2) - (10.8), the term for particle diameter, d_p ,

is substituted with d_{ave} , the average particle diameter as given in Table 10.1. Ar is the Archimedes number given by

$$Ar = \frac{\rho_g(\rho_p - \rho_g)gd_p^3}{\mu_g^2} \quad (10.3)$$

μ_g is gas viscosity (1.81×10^{-5} Pa.s). As for the superficial velocity delineating the core-annulus and fast fluidization regimes, type A choking velocity, V_{ca} , was defined (Bi and Fan, 1991) as;

$$V_{ca} = 21.6\sqrt{gd_p}Ar^{0.105}\left(\frac{G_s}{\rho_g V_{ca}}\right)^{0.542} \quad (10.4)$$

Type A choking velocity was the superficial gas velocity below which particles accumulate at the riser bottom.

The regime map was developed largely based on data for Geldart Group A particles (Bi and Grace, 1995). This study is aimed at finding out if the regime map is applicable to the Geldart Group B particles investigated. Fig. 10.6 presents data from the current work as discrete data points plotted on the regime map demarcated by V_{mp} (Eq. (10.2)) and V_{ca} (Eq. (10.4)) (Bi and Grace, 1995). The x -axis represents the ratio of the overall solids flux to particle density, G_s/ρ_p , while the y -axis the superficial gas velocity, U_g . Because V_{ca} and V_{mp} change with material properties, the three monodisperse particles (Figs. 10.6a - c), the two binary mixtures (Figs. 10.6d– e) and the continuous PSD (Fig. 10.6f) of small glass are plotted separately. The average particle diameter and density for the monodisperse materials and the continuous PSD are given in Table 10.1. The properties for the binary mixtures were the arithmetic averages of the constituents. Each data point represents a particle system at a different operating condition. Where a mixture of upflow and downflow annulus behaviors were observed at different heights, the markers are filled or in bold.

As seen in Fig. 10.6, although none of the three regimes account for upflow annulus, the upflow annulus behaviors observed in the current study are scattered among the regimes.

Two interesting observations can be made. On the regime map of Bi and Grace (1995) the core-annulus regime with the annulus solids downflow is almost non-existent in the cases of large glass (Fig. 10.6b), large HDPE (Fig. 10.6c) and binary-density mixture (Fig. 10.6e). All three particle systems consistently exhibited an upflow annulus regardless of operating conditions. The regime map shows that the fast fluidization regime would be present for the higher G_s conditions and the homogeneous dilute-phase flow regime for the lowest m condition. Secondly, only one data point, which corresponds to the highest solid loading, m , each in monodisperse small glass (Fig. 10.6a) and continuous PSD of small glass (Fig. 10.6f), falls into the core-annulus regime, although both particle systems exhibited a mixture of upflow and downflow annulus at different axial positions in the riser.

Fig. 10.6. Comparison of experimental data with prediction of flow regime map of Bi and Grace (1995) for (a) small glass, (b) large glass, (c) HDPE, (d) binary-size mixture, (e) binary-density mixture, and (f) continuous PSD of small glass. The solid and dotted lines represent V_{mp} (Eq. (10.2)) and V_{ca} (Eq. (10.4)) respectively. ‘Homogeneous dilute-phase flow’ represents upflow annulus with minimal lateral variation of solids concentration, ‘Core-annular flow’ represents a dense downflow annulus and lean upflow core, and ‘Fast fluidization’ represents a dense turbulent bottom and core-annulus top.

Grace et al. (1999) accounted for upflow annulus behavior through the introduction of a new regime called ‘dense suspension upflow’ (DSU). The DSU regime was said to have core-annulus flow with upflow annulus, and solid volume fraction (ϕ) ≥ 0.07 . A semi-empirical correlation was developed to distinguish between conventional core-annulus flow with downflow annulus, termed in the new regime map as ‘fast fluidization’, and the new DSU

regime. The threshold Reynolds number for DSU, Re_{DSU} , and the Archimedes number, Ar , were correlated to give (Grace et al., 1999):

$$Re_{DSU} = 0.113 \left(\frac{G_s d_p}{\mu_g} \right)^{1.192} Ar^{-0.064} \quad (10.5)$$

Analogously, Gupta and Berruti (1998) had published a correlation to distinguish between ‘fast fluidization’ and ‘dense phase flow’ (DPF) regimes. The DPF is similar to DSU in that it requires a core-annulus flow with upflow annulus, but the conditions differ from DSU in that the solids flux has to be greater than $100 \text{ kg/m}^2\text{s}$ and $\phi > 0.15$. The Gupta and Berruti (1998) semi-empirical correlation is expressed as follows

$$Re_{DPF} = 12.55 \left(\frac{G_s}{v_t \rho_p} \right)^{0.55} Ar^{0.36} \quad (10.6)$$

where Re_{DPF} is the threshold Reynolds number for dense phase flow (DPF), and v_t is the particle terminal velocity.

Fig. 10.7 presents data from this study as discrete data points plotted on the regime maps of Grace et al. (1999) (Fig. 10.7a) and Gupta and Berruti (1998) (Fig. 10.7b). The x -axes represent dimensionless groups which are functions of G_s and Ar , and the y -axes represent the particle Reynolds number, Re_p . The lines demarcating the regimes are given by Eqs. (10.5) and (10.6) in Figs. 10.7a and b, respectively. Fig. 10.7 illustrates that, although upflow annulus was observed in most of the cases in this study, all the data points fall in the ‘fast fluidization’ regime (which has a downflow annulus behavior) and none in the DSU or DPF regimes (which predict upflow annulus behavior). Interestingly, the large glass, large HDPE and binary-density systems, which consistently exhibited upflow annulus behavior, are furthest from the DSU and DPF regimes on the regime maps (Fig. 10.7). Therefore, even though the upflow annulus behavior was found in Geldart Group A particle systems, the new regime maps could not account for upflow annulus in CFB risers of Geldart Group B particles.

It should be noted that the regime maps in both, Figs. 10.6 and 10.7, do not incorporate the effect of riser axial position, but rather assign a regime for the entire riser. Hence, this may perhaps be a less than ideal comparison between experimental results, which consider the local annulus flow behavior at different heights along the riser, and the predictions of the regime maps. Nevertheless, the data points in Figs. 10.6 and 10.7 consider the overall operating conditions given in Table 10.2 and the general inadequacy of these established regime maps to predict accurately the annulus flow behavior of Geldart Group B (especially with large d_{ave}) and non-monodisperse particles is clear from the juxtaposition.

Fig. 10.7. Comparison of experimental data with flow regime map of (a) Grace et al. (1999) and (b) Gupta and Berruti (1998). The solid lines represent Eqs. (10.5) and (10.6) in (a) and (b) respectively. Note that ‘Fast fluidization’ represents a dense downflow annulus and lean upflow core (which is unlike Fig. 10.6), while ‘Dense suspension upflow’ (a) and ‘Dense phase flow’ (b) both represent upflow annulus.

Kim et al. (2004) extended the work of Bi and Grace (1995), Grace et al. (1999), and Gupta and Berruti (1998) to propose a new regime map. Three regimes in increasing order of solids velocity were identified: (i) ‘Dense suspension upflow’ with solids upflow across the

entire cross section and $\phi > 0.07$, (ii) ‘Fast fluidization’ with a downflow annulus and an upflow core, and (iii) ‘Dilute pneumatic transport’ with solids upflow across the entire cross-section and $\phi < 0.07$. An important feature of this regime map was the inclusion of the effect of riser axial position by including ϕ in the correlations for regime classification. Two correlations were defined to demarcate the three regimes (Kim et al., 2004). For the demarcation between ‘dense suspension upflow’ and ‘fast fluidization’ regimes:

$$\frac{v_s}{v_t} = 0.004994 \left[\left(\frac{U_g/(1-\phi)}{v_t} \right) \text{Ar}^{0.2} \right]^{1.695} \quad (10.7)$$

And for demarcation between ‘fast fluidization’ and ‘dilute pneumatic transport’ regimes:

$$\frac{v_s}{v_t} = 2540.7 \left[\left(\frac{U_g/(1-\phi)}{v_t} \right) \text{Ar}^{0.2} \right]^{-1.111} \quad (10.8)$$

where v_s is the cross-sectional average particle velocity given by

$$v_s = \frac{G_s}{\rho_p \phi} \quad (10.9)$$

Fig. 10.8 shows the data from the current work as discrete data points plotted on the regime map of Kim et al. (2004). Only the data for the monodisperse systems (small glass, large glass and HDPE) could be plotted here. The solids holdup data for the polydisperse systems (binary mixtures and PSD) were not available. The average cross-sectional solids holdup, ϕ , of the monodisperse systems at each axial position was determined using a combination of pitot tube and extraction probe measurements. Interested readers are referred to a previous publication (Chew et al., 2012c) for detailed measurement and calculation procedures, and the ϕ values obtained thereof. Data points are from different riser axial locations. The filled markers represent downflow annulus and the unfilled ones are for an upflow annulus. The x -axis represents the dimensionless expression $[U_g/(1-\phi)/v_t] \text{Ar}^{0.2}$ and the y -axis represents the dimensionless solids velocity, v_s/v_t . Fig. 10.8 shows that the data points

are scattered in the ‘fast fluidization’ and ‘dense suspension upflow’ regimes, but the classification into the upflow (‘dense suspension upflow’) and downflow (‘fast fluidization’) annulus regimes remains erroneous, thereby highlighting the lack of general applicability to Geldart Group B particles.

Fig. 10.8. Comparison of experimental data with the flow regime map of Kim et al. (2004). The dotted and solid lines represent Eq. (10.7) and Eq. (10.8), respectively. ‘Dilute pneumatic transport’ represents upflow annulus with minimal lateral solids variation, ‘Fast fluidization’ represents a dense downflow annulus and lean upflow core, and ‘Dense suspension upflow’ represents a dense upflow annulus and lean upflow core.

Regime classifications for CFB riser flows were developed from experimental data obtained mostly from Geldart Group A particles and few data from smaller ($d_{ave} \sim 200 \mu\text{m}$) Geldart group B particles. Regime maps developed from Geldart Group B systems and/or polydisperse systems data are lacking. Therefore, expectedly, the predictions of the reported regime maps are closer to experimental observations for small glass rather than large glass and large HDPE. Conversely, despite the properties of small glass being closest to that of Geldart Group A particles than large glass or HDPE, discrepancies between the regime map predictions and experimental data for small glass persisted. This is due to the general lack of Geldart Group B particles data used in the development of the regime maps. Although the discrepancy could be due to the operating conditions and dimensions of the riser being different, the effects of the former are incorporated into the regime maps and the latter fall within the range of values for which the regime maps are applicable. Hence, the limitations of the presently available regime maps underscore the difference between Geldart Group B and the more widely studied Geldart Group A, and the influence of polydispersity.

10.4 Summary of CFB riser studies

The description of the annulus flow behavior of six particle systems (namely, three monodisperse, two binary and the continuous PSD) in a pilot-scale CFB riser under four operating conditions has been laid out above. The corresponding characteristics, including cluster (Chew et al., 2012a, b), mass flux (Chew et al., 2011b), species segregation (Chew et al., 2011a), radial solids concentration (Chew et al., 2012c) and energy of differential pressure signals (Chapter 9), have also been previously reported. Table 10.3 summarizes these observations to provide an overview. Notably, all of these findings have been obtained on the same CFB riser, with the same materials and operating conditions and same analysis instruments. This consistency with riser geometry and analysis instruments is especially

important in view of a recent observation (Cahyadi et al., 2017) that riser configuration and/or the analysis instruments may play more influential roles than are generally attributed to them. Hence, collectively, this comprehensive dataset of riser flow behavior of Geldart Group B particles is expected to be valuable for model development towards enhanced mechanistic understanding.

Table 10.3. Comparison of various riser flow characteristics for the same particle systems, experimental setup and operating conditions.

Characteristic	Monodisperse materials			Binary mixtures		Continuous PSD of glass
	Small glass	Large glass	HDPE	Binary-size	Binary-density	
Cluster appearance probability (Chew et al., 2012a, b)	Most sensitive to riser axial position, negligible with respect to operating condition and material properties.					Most sensitive to operating conditions, more so at the bottom
Cluster duration (Chew et al., 2012a, b)	Most sensitive to axial position, but effect of operating conditions and material properties more pronounced near riser top.			Most sensitive to axial position, but effect of material properties pronounced near riser bottom.		Most sensitive to operating conditions, more so at the top.
	Relatively higher values; increases towards wall.	Relatively lower values; increases towards wall only at $h/H \leq 0.27$; similar profiles at wall at all operating conditions.		Relatively higher values; profiles mimic large glass.	Relatively lower values; profiles mimic neither constituent.	Higher values at the wall near riser bottom.

Characteristic	Monodisperse materials			Binary mixtures		Continuous PSD of glass
	Small glass	Large glass	HDPE	Binary-size	Binary-density	
Cluster frequency (Chew et al., 2012a, b)	Most sensitive to axial position, but effect of operating conditions material properties greatest among the three cluster characteristics (appearance probability, duration, frequency)			Most sensitive to axial position, but effect of material properties and operating conditions pronounced at $h/H \geq 0.47$.		Most sensitive to operating conditions at $h/H \geq 0.47$
	Relatively lower values throughout the riser at wall	Relatively lower values at $h/H < 0.47$ and higher at $h/H \geq 0.47$	Varies depending on h/H , operating condition	Relatively lower values; Profiles mimic small glass	Relatively higher values; Profiles mimic large glass at center, HDPE at wall	At $h/H \geq 0.47$, opposite trends for higher and lower m .
Radial mass flux (Chew et al., 2011b)	Dominant influence of axial position compared to operating conditions and material properties.					
	Dissimilar profiles at lower heights			Profiles similar at $h/H = 0.92$ but different at $h/H \leq 0.27$ at the wall		Profile at lowest m different from other conditions
		Similar flat profiles at $h/H = 0.27-0.47$		Profiles mimic neither constituent	Profiles mimic large glass	
Radial solids concentration (Chew et al., 2012c)	Core-annulus for all operating conditions	Reverse core-annulus at $h/H = 0.92$ for all operating conditions	Reverse core-annulus only for lower G_s .	Not measured due to limitation of pitot tube used.		

Characteristic	Monodisperse materials			Binary mixtures		Continuous PSD of glass
	Small glass	Large glass	HDPE	Binary-size	Binary-density	
Species segregation (Chew et al., 2011a)	<i>Not applicable</i>			More massive species segregate to the wall		
				Relatively more extensive radial and axial segregation		Relatively lesser segregation
				Radial segregation least at $h/H=0.16$		Radial segregation least at $h/H=0.92$
				Composition of larger species increases at the wall with h/H except at lowest m	Composition of denser species at the wall increases with h/H only at lowest m	Composition of larger species at the wall invariant with h/H
				Axial segregation decreases monotonically with increasing m .		Axial segregation non-monotonic with m .
Energy of differential pressure signals (E_{Dj}) (Chapter 9)	Lowest E_{Dj} values	Highest E_{Dj} values	Intermediate E_{Dj} values	Lower E_{Dj} values among the binary systems	Higher E_{Dj} values among the binary systems	<i>Not studied</i>
	Low m gave high E_{dj} ; least sensitive to operating conditions	Low m gave high E_{dj} ; most sensitive to operating conditions	Trends against m disagree with other materials; largest error bars	E_{dj} profiles mimic small glass	E_{dj} profiles mimic large glass	

Characteristic	Monodisperse materials			Binary mixtures		Continuous PSD of glass
	Small glass	Large glass	HDPE	Binary-size	Binary-density	
Annulus flow behavior (this study)	Both upflow and downflow annulus	Upflow annulus		Upflow annulus except at $h/H=0.27$ at highest and second-lowest m	Upflow annulus	Largely downflow annulus, except at lowest m .
		Profiles similar except at $h/H=0.16$ at higher G_s .		Profiles similar at highest m		
	Profiles similar at $h/H \geq 0.47$ at lower G_s and at $h/H=0.16$ and 0.92 at higher G_s .			Large glass affects upflow annulus; profiles dissimilar to constituents.	Profiles similar to both constituents.	Impact of polydispersity seen at lower m .

10.5 Conclusion

The annulus flow behavior of Geldart Group B particles in a pilot-scale CFB riser has been studied. Normalized radial solids flux ($G_{r,net,norm}$) values at the riser wall ($r/R = 0.94$), for six different Geldart group B materials (three monodisperse, two binary mixtures and one PSD), at four different operating conditions and five riser axial positions have been analyzed. The experimental data have been superimposed on the available CFB riser regime maps. These regime maps were largely based on data from Geldart Group A particles.

The followings were the main findings. For the monodisperse materials, (i) particle diameter was more dominant than particle density in determining the solids upflow in the annulus; this was evident from the upflow annulus behavior exhibited by large glass and large HDPE, and the mixture of upflow and downflow annulus behaviors exhibited by small glass; and (ii) the impact of particle density was minimal higher in the riser. For the binary mixtures, (i) both binary mixtures exhibited largely upflow annulus behavior, presumably due to the dominant influence of the larger constituent in dictating upflow annulus behavior; and (ii) the binary-density mixture mimicked the behavior of both its constituent particle species more than that of the binary-size. This suggested that the wall had a more significant effect on the interactions between species of different particle diameters than that of different particle densities. Comparing the monodisperse and continuous PSD of small glass revealed that the impact of polydispersity was more pronounced at lower solid loading (m) conditions. For both monodisperse and binary systems, particle properties had a lesser impact on the annulus flow behavior at the lowest solid loading condition as well as near the riser exit. The available regime maps, developed largely for Geldart Group A particles, were deficient in predicting the experimentally observed annulus flow behavior; hence their applicability was perhaps expectedly limited for the Geldart Group B and non-monodisperse particle systems investigated in this study.

In addition to annulus flow behavior, the key findings of the various riser characteristics (namely, cluster (Chew et al., 2012a, b), mass flux (Chew et al., 2011b), species segregation (Chew et al., 2011a), radial solids concentration (Chew et al., 2012c) and energy of differential pressure signals (Chapter 9)) reported to date on the same system have been summarized in a table here to provide a comprehensive overview.

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Part III

Reactor Modeling

11

Carbon-dioxide methanation in a bubbling fluidized bed - A modeling study

This chapter endeavors to apply a two-phase heterogeneous model to a carbon-dioxide (CO₂) methanation bubbling fluidized bed reactor and assess the effects of varying operating conditions and bed properties on the generation of synthetic natural gas, methane (CH₄).

11.1 Literature review

Increasing amounts of carbon-dioxide (CO₂) in the earth's atmosphere, on one hand, has become a major environmental concern in recent times due to rapid industrialization. Hence, a growing number of efforts are being directed at developing various techniques to effectively capture CO₂ (Rezvani et al., 2009; Wood, 2015). On the other hand, depleting deposits of non-renewable fuels has generated the need to look for renewable sources of energy. The methanation of CO₂ to convert it into Synthetic Natural Gas (SNG) is an effective way of tackling both these problems (Centi and Perathoner, 2009; Centi et al., 2013; Hoekman et al., 2010; Miguel et al., 2015). Along with high combustion efficiency, SNG offers the additional advantages of not requiring to develop any new infrastructure or novel downstream technologies (Chein et al., 2016; Hashimoto et al., 2001). The CO₂ methanation reaction, also known as the Sabatier reaction, which involves passing CO₂ and hydrogen over a metal catalyst has been known since the beginning of the twentieth century (Sabatier and Senderens, 1902):

Though known for a very long time, a standard commercial process has not yet been established for the Sabatier reaction due to issues associated with kinetics, effective catalysts and reactor designing (Schlereth and Hinrichsen, 2014).

A number of studies on developing and assessing the efficacy of various catalysts for reaction (1) have been conducted. Nickel (Ni), ruthenium (Ru) and rhodium (Rh) based catalysts have been popularly investigated (Gnanamani et al., 2014; Wang and Gong, 2010; Wang et al., 2011). The catalyst is prepared by supporting these metals on various materials like zeolites and metal oxides like La_2O_3 , ZrO_2 , CeO_2 , SiO_2 , TiO_2 , Al_2O_3 (Czekaj et al., 2007; Da Silva et al., 2012; Du et al., 2007; Jwa et al., 2013; Kai et al., 1988; Meng et al., 2015; Tada et al., 2012; Zhao et al., 2012). Ni-based catalysts have been most widely examined due to their desirable properties like high methane selectivity, high catalytic activity and low costs (Aziz et al., 2014; Rahmani et al., 2014; Vance and Bartholomew, 1983). Metal composite catalysts like $\text{Ni/CeO}_2\text{-ZrO}_2$ have also been reported (Aldana et al., 2013; Pan et al., 2014; Zhu et al., 2013). Besides Ni, Ru and Rh, palladium (Pd) based catalysts have also been observed to be effective in CO_2 methanation (Park and McFarland, 2009). The kinetics associated with these catalysts have generally been expressed in the Langmuir-Hinshelwood form. One of the early works on kinetics of Ni-based catalysts has been published by Weatherbee and Bartholomew (1982). They investigated an Ni/SiO_2 system and described the kinetics at a pressure of 1.4 bar and a temperature range of 500-600 K. Kai et al. (1988) published expressions for an $\text{Ni-La}_2\text{O}_3$ catalyst. Xu and Froment (1989) published rate expressions for steam reforming, CO_2 methanation and reverse water-gas shift reactions over an $\text{Ni/MgAl}_2\text{O}_4$ catalyst for temperature and pressure ranges of 573-673 K and 3-10 bar respectively.

Comparatively, less amount of work has been done on the reaction engineering and reactor modelling aspects of the Sabatier reaction (Chein et al., 2016; Schlereth and Hinrichsen, 2014). Gao et al. (2012) conducted a detailed thermodynamic analysis of the Sabatier reaction

by the Gibbs free energy minimization method. Brooks et al. (2007) and Hu et al. (2007) developed micro-channel reactors with diameters less than 1 mm for the Sabatier reaction. The former incorporated their kinetic rate equation into a plug-flow reactor model representing the micro-channel reactor. Hwang et al. (2008) investigated a membrane reactor and developed a technique to increase CO₂ conversion. They also developed a model based on numerical methods to assess the performance of their reactor. Sudiro et al. (2010) examined externally cooled multi-tubular reactors to study methanation of a CO₂/CO mixture in the temperature range of 300-550°C. Schlereth and Hinrichsen (2014) conducted a numerical study on the methanation of CO₂ developing homogeneous and heterogeneous models for fixed-bed and membrane reactors and highlighted the importance of temperature control and catalyst activity. Chein et al. (2016) simulated a fixed bed reactor for the CO₂ methanation reaction and investigated the effects of temperature, pressure, feed inlet ratio on CO₂ conversion and compared Ru and Ni based catalysts.

All modelling studies on CO₂ methanation have been carried out on fixed-bed reactors. There have been a few studies (Kopyscinski et al., 2011a, b; Zhang et al., 2015) on CO methanation involving fluidized beds but none on CO₂. Also, a majority of the reactor-modelling studies on the Sabatier reaction focus on its heat removal aspects. But as Kopyscinski et al. (2010) and Chein et al. (2016) have observed, the CO₂ methanation reaction is cognizably sensitive to parameters like temperature, pressure, inlet feed composition etc. Hence, the current study attempts to investigate the effects such parameters on methane production by the Sabatier reaction through a heterogeneous bubbling fluidized bed reactor model.

11.2. Model Description

11.2.1 Fluidized bed

The bubbling fluidized bed investigated here has an inner diameter of 0.5 m and a fluidized bed height of 0.5 m. A two-phase model based on that described by Kopyscinski et al. (2011a) is employed. Fig. 11.1 shows a schematic of the model, which involves mass transfer between the emulsion and bubble phases along the height of the fluidized bed. The following are the principle assumptions:

- The system is comprised of two phases, namely, the bubble phase and the emulsion phase
- The bubble phase is solid-free, and reaction occurs only in the emulsion phase
- The inlet gas enters the reactor at the bottom and goes into either the bubble phase or emulsion phase.
- The two phases interact via mass transfer due both to concentration difference and convection.
- No axial dispersion occurs.
- All gases behave ideally.

Fig. 11.1. Schematic of the two-phase model (Kopyscinski et al., 2011a) for the bubbling fluidized bed methanation reactor. The bed height is 0.5 m, $y_{e,i}$ and $y_{b,i}$ are respectively the emulsion and bubble phase concentrations (mol m^{-3}) of species i , z is the axial position along the fluidized bed column (m), $K_{G,i}$ is the inter-phase mass transfer (m s^{-1}) of species i due to concentration gradient and N_{vc} is the convective inter-phase mass transfer ($\text{mol m}^{-1} \text{s}^{-1}$).

The mass balance for species i for the bubble phase is given by

$$\frac{dy_{b,i}}{dz} = -[K_{G,i}aA(y_{b,i} - y_{e,i}) + N_{vc}x_{b,i}]\frac{z}{VU} \quad (11.2)$$

where y denotes the concentration (mol m^{-3}) of component i , the subscripts b and e represent respectively the bubble and emulsion phases, z is the height along the fluidized bed column (m), $x_{b,i}$ is the mole fraction of component i in the bubble phase (-), a is the specific mass-transfer area (m^{-1}), A is the cross-sectional area of the reactor (m^2), $K_{G,i}$ is the mass-transfer coefficient (m s^{-1}), N_{vc} is the total bulk flow from the bubble phase to the emulsion phase (mol

$\text{m}^{-1} \text{s}^{-1}$), V is the total volume of the reactor (m^3) and U is the superficial gas velocity (m s^{-1}).

The mass balance for species i for the emulsion phase is given by

$$\frac{dy_{e,i}}{dz} = -[K_{G,i}aA(y_{b,i} - y_{e,i}) + N_{vc}x_{b,i} + (1 - \varepsilon_b)(1 - \varepsilon_{mf})\rho_p AR_i] \frac{z}{VU} \quad (11.3)$$

where $(1 - \varepsilon_b)$ is the volume fraction of the emulsion phase, $(1 - \varepsilon_{mf})$ is the volume fraction of the particles at the minimum fluidization velocity, ρ_p is the particle density and R_i is the rate of consumption or formation of component i . The convection-induced flow is given by

$$N_{vc} = \sum_i K_{G,i}aA(y_{b,i} - y_{e,i}) + (1 - \varepsilon_b)(1 - \varepsilon_{mf})\rho_p A \sum_i R_i \quad (\text{mol m}^{-1} \text{s}^{-1}) \quad (11.4)$$

where R_i is the product of the stoichiometric factor of species i , v_i , and reaction rate, r . Hence,

$$R_{CO_2} = -r \quad (11.5)$$

$$R_{H_2} = -4r \quad (11.6)$$

$$R_{CH_4} = r \quad (11.7)$$

$$R_{H_2O} = 2r \quad (11.8)$$

The following boundary conditions are applied at $z = 0$,

$$y_{b,i}|_{z=0} = y_{b,i,feed} \quad (11.9)$$

$$y_{e,i}|_{z=0} = y_{e,i,feed} \quad (11.10)$$

11.2.2 Hydrodynamics

Table 11.1 lists all the correlations used in this study to determine various hydrodynamic parameters for the bubbling fluidized bed. The disadvantage of using these correlations is that they are empirical or semi-empirical and will inevitably give best predictions within the scope of the systems from which they were developed. However, the use

of such correlations was unavoidable, since a purely physically-based model describing the hydrodynamics of a bubbling fluidized is not yet available. Nevertheless, the two most important correlations to the sensitivity analysis in the next section, those for the bubble diameter (d_b) and the minimum fluidization velocity (U_{mf}), were selected based on the following factors: (i) the gas concentration profiles calculated using in the model the selected correlation for d_b , given by Werther (1976) was found by Kopyscinski et al. (2011b) to best fit the measured experimental data in CO bubbling fluidized beds over four other correlations (Darton et al., 1977; Hillgardt and Werther, 1987; Mori and Wen, 1975; Rowe, 1976) and (ii) the Ergun (1952) correlation for small particles i.e. the selected U_{mf} correlation includes the parameters of particle sphericity (ϕ) and bed voidage at minimum fluidization (ε_{mf}), which are a part of the sensitivity analysis and is the correlation from which the most popular U_{mf} correlation by Wen and Yu (1966) is developed. Note that the Wen and Yu (1966) correlation was not selected since it does not explicitly include ϕ and ε_{mf} .

Table 11.1. Correlations for hydrodynamic parameters

Parameters	Correlations	Units	Eqn. No.	Refs.
Bubble diameter	$d_b = d_{b0} \left[1 + 27.2(U - U_{mf}) \right]^{\frac{1}{3}} (1 + 6.84z)^{1.21},$ <p>where d_{b0} is the initial bubble diameter, given as 0.00853 m; U is the superficial gas velocity (m s^{-1}); U_{mf} is the minimum fluidization velocity (m s^{-1}); z is the height along the bubbling fluidized bed column (m)</p>	m	(11.11)	Werther (1976)

Parameters	Correlations	Units	Eqn. No.	Refs.
Bubble velocity	$U_b = 0.69(U - U_{mf}) + 0.4479\sqrt{gd_b}$ <p>where g is gravitational acceleration (9.81 m s^{-2})</p>	m/s	(11.12)	Hilligardt and Werther (1985); Hilligardt and Werther (1986); Kopyscinski et al. (2011a)
Bubble phase holdup	$\varepsilon_b = \frac{v_b}{u_b}$ <p>where v_b is the visible bubble phase velocity based on tube diameter (m s^{-1})</p>	-	(11.13)	Kopyscinski et al. (2011a)
Specific mass-transfer area	$a = \frac{6\varepsilon_b}{d_b}$	m^{-1}	(11.14)	Kopyscinski et al. (2011a)
Viscosity of gas mixture	$\mu = \frac{\sum_i x_i \mu_i}{\sum_i x_i \phi_{ij}}$ $\phi_{ij} = \frac{\left[1 + \left(\frac{\mu_i}{\mu_j}\right)^{0.5} \left(\frac{M_j}{M_i}\right)^{0.25}\right]^2}{\left[8 \left(1 + \frac{M_i}{M_j}\right)\right]^{0.5}}$ <p>where subscripts i and j represent species i and j, respectively; x is mole fraction (-), μ is viscosity (Pa s); and M is molecular weight (kg mol^{-1}).</p>	Pa.s	(11.15)	Wilke (1950)
Density of gas mixture	$\rho = \sum_i x_i \rho_i$ <p>where ρ_i is the density of component i (kg m^{-3})</p>	kg/m^3	(11.16)	

Parameters	Correlations	Units	Eqn. No.	Refs.
Minimum fluidization velocity	$U_{mf} = \frac{d_p^2 (\rho_p - \rho) g}{150 \mu} \frac{\varepsilon_{mf}^3 \varphi^2}{(1 - \varepsilon_{mf})}$ <p>where d_p is particle diameter (m), ρ_p is particle density (kg m^{-3}), ρ is gas density (kg m^{-3}), ε_{mf} is bed voidage at minimum fluidization (-), φ is particle sphericity (-)</p>	m/s	(11.17)	Ergun (1952) in Kunii and Levenspiel (1991)
Binary diffusion coefficient of species i and j	$D_{i,j} = 0.01013 \frac{T^{1.75} [(1/M_i) + (1/M_j)]^{1/2}}{P \left[(\sum v_i)^{1/3} + (\sum v_j)^{1/3} \right]^2}$ <p>where T is temperature (K), P is pressure (Pa), v_i, v_j are the atomic diffusion volumes (Fuller et al., 1969; Fuller et al., 1966) ($\text{m}^3 \text{mol}^{-1}$)</p>	m^2/s	(11.18)	Fuller et al. (1969); Fuller et al. (1966)
Molecular diffusion coefficient of species i	$D_{i,mix} = \frac{1 - x_i}{\sum_j \left(\frac{x_j}{D_{i,j}} \right)}$	m^2/s	(11.19)	Fuller et al. (1969); Fuller et al. (1966)
Mass-transfer coefficient	$K_{G,i} = \frac{u_{mf}}{3} + \sqrt{\frac{4 D_{i,mix} \varepsilon_{mf} U_b}{\pi d_b}}$	m/s	(11.20)	Sit and Grace (1981)

11.2.3 Kinetics

The kinetics of CO_2 methanation for an integral reactor (Doraiswamy and Tajbl, 1974) developed by Kai et al. Kai et al. (1988) are adopted in this study, since CO_2 methanation has not been experimentally studied in a bubbling fluidized bed reactor. The rate of methanation is given by a Langmuir-Hinshelwood type of expression (Kai et al., 1988):

$$r_{CH_4} (\text{mol kg}^{-1} \text{s}^{-1}) = \frac{k P_{H_2}^{\frac{1}{2}} P_{CO_2}^{\frac{1}{3}}}{\left(1 + K_{H_2} P_{H_2}^{\frac{1}{2}} + K_{CO_2} P_{CO_2}^{\frac{1}{3}} + K_{H_2O} P_{H_2O} \right)^2} \quad (11.21)$$

The dependence of reaction constant, k , on temperature, T , was obtained from an Arrhenius plot (Kai et al., 1988).

$$k \text{ (mol kg}^{-1}\text{s}^{-1}\text{kPa}^{-5/6}\text{)} = 9297.21e^{\left(\frac{-72500}{RT}\right)} \quad (11.22)$$

The constants K_{H_2} , K_{CO_2} and K_{H_2O} are obtained via the van't-Hoff equation.

$$K_{H_2} \text{ (kPa}^{-1/2}\text{)} = 3.77 \cdot 10^{-11}e^{\left(\frac{90200}{RT}\right)} \quad (11.23)$$

$$K_{CO_2} \text{ (kPa}^{-1/2}\text{)} = 1.43 \cdot 10^{-4}e^{\left(\frac{29500}{RT}\right)} \quad (11.24)$$

$$K_{H_2O} \text{ (kPa}^{-1}\text{)} = 2.75 \cdot 10^{-8}e^{\left(\frac{964300}{RT}\right)} \quad (11.25)$$

Partial pressures (Pa) are converted to units of concentration (mol/m³) by

$$y_{e,i} = \frac{P_i RT}{1000} \quad (11.26)$$

Only concentrations of the emulsion phase are considered as the reaction occurs only in the emulsion phase.

11.3 Concentration profiles

Fig. 11.2 shows the concentration profiles of the various species (namely, reactants H_2 and CO_2 , products CH_4 and H_2O , and carrier gas N_2) in both the emulsion and bubble phases along the reactor height. The solid lines represent the emulsion phase while the dashed lines represent the bubble phase. Note that the reaction takes place only in the emulsion phase and the presence of product components in the bubble phase is a result of the mass transfer between the two phases. Table 11.2 lists the base case parameters used to generate the profiles in Fig. 11.2. Six key observations from Fig. 11.2 are: (i) all profiles are expectedly non-linear in accordance with Langmuir-Hinshelwood kinetics; (ii) the rates of depletion and accumulation

of reactants and products respectively decrease with increasing column height, indicating that the reaction decelerates with height; (iii) the concentration of H_2O in both phases is twice that of CH_4 , in accordance to the stoichiometric relationship given in Eq. (11.1); (iv) the concentration of the inert N_2 expectedly remains constant throughout the bed in both phases; (v) generally, the concentrations of the reactants (namely, H_2 and CO_2) are higher in the bubble phase than emulsion phase, whereas the concentrations of the products (namely, CH_4 and H_2O) are higher in the emulsion phase than bubble phase, indicating that the inter-phase mass transfer is limiting; and (vi) the concentrations of the reactants (namely, H_2 and CO_2) in the bubble phase increases slightly at the bottom of the reactor ($z \approx 0$), which may be attributed to the slight decrease in the bubble diameter, as has been noted previously in CO methanation fluidized beds (Kopyscinski et al., 2011a).

Table 11.2. Base case parameters for results in Fig. 11.2

Parameter	Value	Unit
Temperature	570	K
Pressure	2	atm
Column diameter	0.5	m
Bed height	0.5	m
Bed voidage at minimum fluidization, ε_{mf}	0.42	-
CO_2/H_2 inlet ratio	4:1	-
Inert gas (N_2) mole fraction	0.2	-
Ratio of inlet gas velocity to minimum fluidization velocity, (U/U_{mf})	1.5	-
Particle diameter, d_p	400	μm
Particle density, ρ_p	4000	kg m^{-3}
Particle sphericity, ϕ	1	-

Fig. 11.2. Concentration profiles of reactants and products based on the conditions given in Table 11.2. The values of the parameters used are listed in Table 11.2.

11.4 Sensitivity analysis

Effects of temperature, inlet superficial gas velocity, particle diameter, particle density, particle sphericity and bed voidage at minimum fluidization are analyzed through Sections 11.4.1 – 11.4.6 respectively. The effects of varying pressure and $\text{H}_2\text{:CO}_2$ inlet ratio are as expected and are recounted in Sections 11.4.7 and 11.4.8 respectively.

11.4.1 Effect of temperature

Fig. 11.3 shows the concentration profiles along the reactor height (z) at four different temperatures (namely, 510 K, 540 K, 570 K and 600 K). The lower and upper bounds of temperature were chosen in accordance with the temperature range (513 K – 593 K) for which the experiments to determine the kinetics (Eq. (11.22)) were carried out (Kai et al., 1988). Each

sub-plot represent each reactant (i.e., CO₂ or H₂) or product (i.e., CH₄ or H₂O). Other than temperature, the parameters are as given in Table 11.2. The solid lines represent the emulsion phase while the dashed lines represent the bubble phase. Fig. 11.4 further shows the outlet (i.e., at $z = 0.5$ m) concentrations of CH₄ as a function of temperature. The following two observations are clear from Figs. 11.3 and 11.4. Firstly, the concentrations of the reactants (Fig. 11.3a and c) generally decrease while the concentrations of the products (Fig. 11.3b and d) increase as temperature increases up to 570 K, beyond which the effect of temperature diminishes. This is due to the complete depletion of the reactants by $z = 0.2$ at the highest temperature of 600 K. Fig. 11.4 further shows that the concentration of CH₄ particularly in the emulsion phase actually decreases slightly with temperature between 580 and 600 K. This agrees with previous CO₂ methanation results in a fixed bed reactor (Chein et al., 2016), whereby the reaction rate was also observed to increase with temperature up to a certain point before decreasing. Secondly, Fig. 11.3 shows that the concentrations in the emulsion and bubble phases become more similar higher along the bed (i.e., $z > 0.35$) at the highest temperature of 600 K, while Fig. 11.4 further affirms that the outlet CH₄ concentration does become similar for the higher temperatures of 590 – 600 K. This indicates that the concentrations in both phases equilibrate to the same values in the absence of reactions. Notably, despite the depletion of the reactants by $z = 0.2$, the concentrations of products (Fig. 11.3b and d) in the two phases do not equate until $z = 0.5$.

Fig. 11.3. Concentration profiles of (a) CO_2 , (b) CH_4 , (c) H_2 and (d) H_2O versus reactor height (z) at four different temperatures (namely, 510 K, 530 K, 570 K and 600 K). The solid lines represent the emulsion phase while the dashed lines represent the bubble phase.

Fig. 11.4. Outlet (i.e., $z = 0.5$ m) concentration of the emulsion phase (solid line) and bubble phase (dashed line) of CH₄ against temperature

To better understand the effect of temperature, Fig. 11.5a shows the rate of the methanation reaction against the reactor height (z) at the four temperatures investigated in Fig. 11.3. Interestingly, as temperature increases, the reaction rate increases at the bottom of the reactor but decreases at the top. Hence, the higher the temperature is, the steeper the profile becomes. The reaction rate decreases with temperature at higher heights due to the faster depletion of the reactants (Eq. (11.21)). Fig 11.5b, which displays the reaction rate at the reactor inlet ($z = 0$) against temperature, confirms this. As seen from Fig. 11.5b, the reaction rate at $z = 0$ increases monotonically with temperature, indicating that the kinetics favor higher temperature but are limited by the depleting amounts of reactants higher up in the reactor with increasing temperature. In other words, if the exhausting reactants were to be continually

replenished as consumed throughout the reactor column, the reaction rate would be greater at higher temperatures throughout the reactor.

Fig. 11.5. (a) Reaction rate versus reactor height (z) at the four temperatures (namely, 510 K, 530 K, 570 K and 600 K) investigated in Fig. 11.3 and (b) Reaction rate at reactor inlet ($z = 0$) versus temperature.

11.4.2 Effect of inlet flow velocity

Fig. 11.6 shows the concentration profiles of the reactants and products along the reactor height at four different inlet superficial gas velocities (U) in terms of multiples of the minimum fluidization velocity (namely, $U = 1.1U_{mf}$, $1.5U_{mf}$, $2U_{mf}$ and $3U_{mf}$), while Fig. 11.7 shows the outlet concentrations of CH_4 in both phases with respect to the inlet superficial gas velocity (U). Other than U , all parameters are as given in Table 11.2. Four important highlights from Figs. 11.6 and 11.7 are as follows: (i) For the emulsion phase, the CH_4 outlet concentration decreases with increasing inlet flow velocity, (ii) The rate of decrease in the outlet CH_4 concentration in the emulsion phase increases with increasing inlet flow velocity, indicating that the CH_4 yield is more sensitive to inlet flow at higher rather than lower values, (iii) For the

bubble phase the CH₄ outlet concentration varies non-monotonically with the inlet flow velocity, namely increases and decreases, and (iv) Higher in the reactor, the emulsion phase concentration profiles of the reactants and products are flatter at lower inlet flow velocities. Regarding (i), it has been observed in CO methanation fluidized beds that the fluidization quality reduces as inlet velocity increases since mass transfer between the two phases is hampered (Chavarie and Grace, 1976). Also, the higher superficial gas velocity causes a decrease in the residence time of the inlet gas mixture and significant gas bypass (Huilin et al., 2007; Limtrakul et al., 2003; Limtrakul et al., 2005) resulting in a lower outlet product concentration. The non-monotonic trend in (iii) of the outlet CH₄ concentration with respect to inlet velocity observed in the bubble phase can be attributed to the bubble phase hold-up which is a function of the difference between the inlet velocity and the minimum fluidization velocity, $(U - U_{mf})$. $(U - U_{mf})$ can vary from <1 to >1 , which results in the non-monotonic trend. The increased gas bypass at higher inlet velocities mentioned earlier also explains the higher outlet reactant concentrations at $U = 3U_{mf}$ and $U = 3U_{mf}$ (Fig. 11.6 a and c) and the flatter concentration profiles of the reactants and products observed in (iv).

Fig. 11.6. Concentration profiles of (a) CO_2 , (b) CH_4 , (c) H_2 and (d) H_2O versus reactor height (z) at four different inlet flow velocities (namely, $U = 1.1U_{mf}$, $U = 1.5U_{mf}$, $U = 2U_{mf}$ and $U = 3U_{mf}$). The solid lines represent the emulsion phase while the dashed lines represent the bubble phase.

Fig. 11.7. Outlet (i.e., $z = 0.5$ m) concentration of the emulsion phase (solid red line) and bubble phase (dashed blue line) of CH_4 against the ratio of inlet flow velocity (U) to the minimum fluidization velocity (U_{mf}).

11.4.3 Effect of particle diameter

Fig. 11.8 shows the concentration profiles of the reactants and products along the reactor at four different particle diameters ($d_p = 150 \mu\text{m}$, $d_p = 400 \mu\text{m}$, $d_p = 600 \mu\text{m}$ and $d_p = 750 \mu\text{m}$), while Fig. 11.9 shows the emulsion phase and bubble outlet concentrations of CH_4 varying against d_p . All other parameters are as given in Table 11.2. As can be seen from Figs. 11.8 and 11.9, the CH_4 outlet concentration decreases with increasing d_p for the emulsion phase. This can be attributed to the increasing surface area of particles with smaller d_p , which provide a larger surface for the reaction to take place. Accordingly, the bubble phase also exhibits the same trend with respect to d_p as the emulsion phase. The only exception is at the lowest d_p ($150 \mu\text{m}$) where the outlet CH_4 concentration is slightly lower than that at $d_p = 200 \mu\text{m}$ (Fig. 11.9). This could be attributed to the fact that the $150 \mu\text{m}$ particles are near the transition of Geldart

Group A to B. It has been previously observed (Zhang et al., 2015) that the mass transfer between the emulsion and bubble phases is poorer for Geldart Group A than Group B particles, which may be reason for the reduced CH₄ outlet concentration in the bubble phase for the 150 μm particles. Two further observations are noteworthy: (i) Particle diameter hardly affects the outlet CH₄ concentration for $d_p < 300\text{ }\mu\text{m}$ (Fig. 11.9) and (ii) For the 150 μm particles, the concentrations of the reactants and products remain constant from a height of 0.7 m along the reactor (Fig. 11.8). These two observations indicate that though the reaction decelerates with increasing d_p , it does not affect the methane yield markedly until the catalyst particles are bigger than 300 μm.

Fig. 11.8. Concentration profiles of (a) CO_2 , (b) CH_4 , (c) H_2 and (d) H_2O versus reactor height (z) at four different particle diameters (namely, $d_p = 150 \mu\text{m}$, $d_p = 400 \mu\text{m}$, $d_p = 600 \mu\text{m}$ and $d_p = 750 \mu\text{m}$). The solid lines represent the emulsion phase while the dashed lines represent the bubble phase.

Fig. 11.9. Outlet (i.e., $z = 0.5$ m) concentration of the emulsion phase (solid red line) and bubble phase (dashed blue line) of CH₄ against particle diameter, d_p .

11.4.4 Effect of particle density

Fig. 11.10 shows the concentration profiles of the reactants and products along the reactor at four different particle densities ($\rho_p = 2000 \text{ kg/m}^3$, $\rho_p = 3000 \text{ kg/m}^3$, $\rho_p = 4000 \text{ kg/m}^3$ and $\rho_p = 5000 \text{ kg/m}^3$). All other parameters are as given in Table 11.2. As can be seen from Fig. 11.10, both, the emulsion and bubble phase concentration curves at all four ρ_p values almost overlap. While the bubble phase concentration curves are almost concurrent, the emulsion phase concentration curves are placed slightly higher for products and slightly lower for reactants with increasing ρ_p , indicating that the reaction rates are slightly higher for less dense particles. The maximum difference between the highest (least dense particles) and lowest (most dense particles) outlet CH_4 concentrations is, however, less than 8% (Fig. 11.10e). Since the effect of ρ_p on the system is far lesser than that of the other parameters examined here, no further investigation was carried out.

Fig. 11.10. Concentration profiles of (a) CO_2 , (b) CH_4 , (c) H_2 and (d) H_2O versus reactor height (z) at four different particle densities (namely, $\rho_p = 2000 \text{ kg/m}^3$, $\rho_p = 3000 \text{ kg/m}^3$, $\rho_p = 4000 \text{ kg/m}^3$ and $\rho_p = 5000 \text{ kg/m}^3$), and (e) Fig. 11.10a zoomed-in (fuchsia rectangle) for better distinguishing the trends. The solid lines represent the emulsion phase while the dashed lines represent the bubble phase.

11.4.5 Effect of particle sphericity

Fig. 11.11 shows the concentration profiles of the reactants and products along the reactor at four different particle sphericities ($\phi = 0.6$, $\phi = 0.75$, $\phi = 0.85$ and $\phi = 1$), while Fig. 11.12 shows the emulsion phase and bubble outlet concentrations of CH_4 varying against ϕ . All other parameters are as given in Table 11.2. It can be seen from Fig. 11.11 that the concentration trends of the reactant and products in the emulsion phase reach a constant value higher up in the reactor with increasing ϕ , though the outlet concentrations are almost constant. This indicates a faster reaction rate for beds with low sphericity particles. The bubble phase concentration trends are similar to their emulsion phase counterparts, but the outlet concentrations reveal a non-monotonic relationship with respect to ϕ , which is discussed in the following sentences on the basis of Fig. 11.12. Fig. 11.12 reveals a non-monotonic relationship, namely increasing and decreasing, with a peak at $\phi = 0.55$, between the outlet CH_4 concentration and ϕ for both, the emulsion and bubble phases. The non-monotonicity is pronounced for the bubble phase, while for the emulsion phase the outlet CH_4 concentration increases very gradually with respect to ϕ until $\phi = 0.55$ and decreases relatively rapidly for $\phi > 0.55$. The following hypothesis can be proposed for these observations. Particles with low ϕ have been observed to fluidize relatively poorly compared to more spherical particles (Puspasari et al., 2013). This hampers the interaction between the bubble and emulsion phases (Zhang et al., 2015), which results in poor mass transfer from the emulsion to the bubble phase. On the other hand, at high ϕ values, the fast-moving larger bubbles will reduce the inter-phase mass transfer resulting in lower outlet CH_4 concentrations in the bubble phase. In the emulsion phase, more spherical particles possessing lower surface areas than irregular particles with lower ϕ values, will result in a lower outlet CH_4 concentrations for the former. However, the poor quality of fluidization and gas bypass in the case of highly non-spherical particles ($\phi <$

0.55), would compensate for the higher surface area resulting in an almost constant outlet CH_4 concentration for $\varphi < 0.55$ in the emulsion phase.

Fig. 11.11. Concentration profiles of (a) CO_2 , (b) CH_4 , (c) H_2 and (d) H_2O versus reactor height (z) at four different particle sphericities (namely, $\varphi = 0.6$, $\varphi = 0.75$, $\varphi = 0.85$ and $\varphi = 1$). The solid lines represent the emulsion phase while the dashed lines represent the bubble phase.

Fig. 11.12. Outlet (i.e., $z = 0.5$ m) concentration of the emulsion phase (solid red line) and bubble phase (dashed blue line) of CH₄ against particle sphericity (ϕ)

11.4.6 Effect of bed voidage at minimum fluidization

Fig. 11.13 shows the concentration profiles of the reactants and products along the reactor at four different bed voidages at minimum fluidization ($\varepsilon_{mf} = 0.35$, $\varepsilon_{mf} = 0.40$, $\varepsilon_{mf} = 0.45$ and $\varepsilon_{mf} = 0.50$), while Fig. 11.14 shows the emulsion phase and bubble outlet concentrations of CH₄ varying against ε_{mf} . All other parameters are as given in Table 11.2. As can be seen from Fig. 11.13, the concentration curves are placed higher in case of products and lower in case of reactants with decreasing ε_{mf} , indicating that the reaction accelerates at lower ε_{mf} values. Fig. 11.14 reveals that the outlet CH₄ concentration in both phases is almost unaffected until $\varepsilon_{mf} = 0.4$, where after it decreases with ε_{mf} , reflecting the decreased amount of catalyst available per unit volume in the bed. A closer look at the figure (inset in Fig. 11.14) shows a slight non-monotonic trend, namely increasing then decreasing of the CH₄ outlet emulsion and consequently bubble (not shown in inset) phase concentrations. ε_{mf} affects a host of parameters

in the fluidized bed system, like the U_{mf} , bubble diameter, bubble velocity, mass transfer coefficients and the inter-phase bulk flow, which in turn affect the outlet concentrations. An interplay of these factors could be responsible for the outlet concentration trends observed with respect to ε_{mf} . A detailed analysis of these factors vis-à-vis ε_{mf} and the outlet concentrations is, however, out of the scope of this work, however open to future investigation.

Fig. 11.13. Concentration profiles of (a) CO_2 , (b) CH_4 , (c) H_2 and (d) H_2O versus reactor height (z) at four different bed voidages at minimum fluidization (namely, $\varepsilon_{mf} = 0.35$, $\varepsilon_{mf} = 0.40$, $\varepsilon_{mf} = 0.45$ and $\varepsilon_{mf} = 0.50$). The solid lines represent the emulsion phase while the dashed lines represent the bubble phase.

Fig. 11.14. Outlet (i.e., $z = 0.5$ m) concentration of the emulsion phase (solid red line) and bubble phase (dashed blue line) of CH₄ against bed voidage at minimum fluidization (ϵ_{mf}). The inset shows the emulsion phase curve of Fig. 11.14 zoomed in from $\epsilon_{mf} = 0.3$ to 0.36, which reveals a slight non-monotonic trend.

11.4.7 Effect of pressure

Fig. 11.15 shows the concentration profiles of the reactants and products along the reactor height at four different pressures (namely, 1 atm, 2 atm, 4 atm and 8 atm). Other than pressure, the parameters used are given in Table 11.2. Moreover, Fig. 11.16 shows the outlet (i.e., $z = 0.5$ m) concentrations of CH₄ in both phases against pressure. Figs. 11.15 and 11.16 collectively reveal the following. Firstly, the concentrations of both the reactants and products increase with pressure, which agrees with previous studies on CO methanation (Zhang et al., 2015). Secondly, the outlet concentrations of the reactants are non-zero for 4 atm and 8 atm pressures indicating that they were not fully utilized for the reaction and the bed height of 0.5 m is not sufficient for the reaction to complete. This is reflected in the profiles of the products

where their concentration profiles are flatter at the lower pressures of 1 atm and 2 atm, whereas the slopes of the profiles increase for the pressures of 4 atm and 8 atm. Thirdly, it can be seen from Fig. 11.16 that the rate of increase of outlet CH₄ concentration decreases with increasing pressure. This is again related to the previous observation in that more bed inventory is required for the reaction to complete at higher pressures, hence, the outlet concentration i.e. concentration at a column height of 0.5 m increases by smaller amounts as pressure increases. Fourthly, the difference between the CH₄ outlet concentrations of the emulsion and bubble phases increases with increasing pressure. Specifically, at 1 atm, the CH₄ outlet concentration at 1 atm is about 3.5 mol/m³ for both phases whereas that at 8 atm is about 17.5 mol/m³ for the emulsion phase and 13.5 mol/m³ for the bubble phase. This indicates that as pressure increases, the rate of production of CH₄ outpaces its rate of transport from the emulsion to the bubble phase. Recall here that the reaction takes place only in the emulsion phase, hence CH₄ transport is from the emulsion to the bubble phase only.

Fig. 11.15. Concentration profiles of (a) CO_2 , (b) CH_4 , (c) H_2 and (d) H_2O versus reactor height (z) at four different pressures (namely, 1 atm, 2 atm, 4 atm and 8 atm). The solid lines represent the emulsion phase while the dashed lines represent the bubble phase.

Fig. 11.16. Outlet (i.e., $z = 0.5$ m) concentration of the emulsion phase (solid red line) and bubble phase (dashed blue line) of CH₄ against pressure.

11.4.8 Effect of inlet molar ratio of H₂:CO₂

Fig. 11.17 shows the concentration profiles of the reactants and products along the reactor height at four different inlet molar ratios of H₂:CO₂ (namely, 3:1, 4:1, 5:1 and 6:1], while Fig. 11.18 shows the outlet concentrations of CH₄ in both phases with respect to the ratios. Other than the molar ratios of H₂:CO₂, all parameters are as given in Table 11.2. It should be noted that the reaction (Eq. (11.1)) stipulates that the stoichiometric ratio of H₂:CO₂ is 4:1. Four noteworthy observations from Figs. 11.17 and 11.18 are as follows. Firstly, Fig. 11.18 expectedly shows that the peak CH₄ concentration is obtained when the inlet molar ratio of H₂:CO₂ is 4 (Eq. (11.1)). This agrees with previous observations of CO₂ methanation in a fixed-bed reactor (Chein et al., 2016). Secondly, the product concentrations (Fig. 11.17b and 11.17d) are not affected by the inlet molar ratios lower in the bed ($z < 0.1$ m), but become so once one

of the two reactants become depleted, then diverge according to the inlet $\text{H}_2:\text{CO}_2$ values, indicating that the shortage of either reactant when fed at a non-stoichiometric ratio (inlet $\text{H}_2:\text{CO}_2 \neq 4$) starts affecting CH_4 yield at about a fifth way through the total bed height. Thirdly, the difference between CH_4 outlet concentrations in the emulsion and bubble phases varies non-monotonically with inlet $\text{H}_2:\text{CO}_2$ like the concentrations themselves, indicating that the rate of production of CH_4 in the emulsion phase dominates the mass transfer to the bubble phase more as inlet $\text{H}_2:\text{CO}_2$ approaches the stoichiometric ratio. Fourthly, the CH_4 outlet concentration with respect to inlet $\text{H}_2:\text{CO}_2$ in both phases exhibits a convex trend at inlet $\text{H}_2:\text{CO}_2 < 4$ and a concave trend at inlet $\text{H}_2:\text{CO}_2 > 4$ indicating that when the reactant feed is near the stoichiometric ratio, a shortage of CO_2 affects CH_4 outlet concentration more than that of H_2 .

Fig. 11.17. Concentration profiles of (a) CO₂, (b) CH₄, (c) H₂ and (d) H₂O versus reactor height (z) at four different H₂:CO₂ inlet molar ratios [namely, 3:1, 4:1, 5:1 and 6:1]. The solid lines represent the emulsion phase while the dashed lines represent the bubble phase.

Fig. 11.18. Outlet (i.e., $z = 0.5$ m) concentration of the emulsion phase (solid red line) and bubble phase (dashed blue line) of CH_4 against inlet molar ratio of $H_2:CO_2$.

11.5 Conclusion

A two-phase heterogeneous model was applied to a bubbling fluidized bed carbon-dioxide methanation reactor to assess the effects of various operating conditions, namely, temperature, pressure, inlet flow velocity and reactant ratio; and bed properties, namely, particle diameter, density and sphericity, and bed voidage at minimum fluidization on the production of methane.

The modelling results reveal the following: (i) Outlet CH_4 concentration exhibited a non-monotonic trend with respect to temperature, namely increased rapidly, then decreased gradually, owing to the reverse-reaction becoming more dominant at higher temperatures; (ii) Outlet CH_4 concentration increases with increasing pressure, however more bed inventory would be required for the reaction to complete at higher pressures; (iii) Outlet CH_4

concentration decreases with increasing inlet flow velocity due to increased gas bypass; (iv) With respect to the H_2/CO_2 inlet, a peak concentration is obtained at the stoichiometric reactant ratio, namely 4:1; (v) The reaction accelerates as the bed particle diameter (d_p) decreases, however the outlet CH_4 concentration remains generally unaffected till a d_p value of 300 μm , where after it decreases with increasing d_p ; (vi) Outlet CH_4 concentration is virtually unaffected by particle density, ρ_p ; (vii) The reaction accelerates for particles with decreasing sphericity, ϕ , while the outlet CH_4 concentration displays a non-monotonic trend, namely increasing very gradually, then decreasing with ϕ ; and (viii) With respect to the voidage at minimum fluidization, ε_{mf} , the outlet CH_4 concentration almost remains unaffected with increasing ε_{mf} till $\varepsilon_{mf} = 0.4$ and decreases thereafter.

This study thus throws light on the various parameters affecting CO_2 methanation (Sabatier reaction) in a fluidized bed. Experimental data for the Sabatier reaction in a fluidized bed is unfortunately not readily available, due to which the results of this modeling study could not be validated against experiment. The next step in progression will hence be to direct efforts towards gathering such data. The current study would provide a basis for experimental work, which would in turn be used to validate this model.

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12

Conclusion and future perspectives

This thesis presented the research efforts aimed at improving the physical understanding of two very important phenomena in gas-solid flow, namely, pneumatic conveying and fluidization. In particular, studies focused on the minimum pickup velocity (U_{pu}), defined as the minimum gas velocity required to initiate motion of a particle initially at rest (Zenz, 1964), a fundamental parameter in pneumatic conveying; the minimum fluidization velocity, defined as the superficial gas velocity at which the drag force of the upward moving gas is equal to the apparent weight of the particle bed (Fan and Zhu, 1998; Kunii and Levenspiel, 1991), analyzing pressure signals and solid flow in circulating fluidized bed (CFB) risers and fluidized bed reactor modelling. The objective was to assess the effects of particle properties and operating conditions. Key takeaways from specific studies undertaken are summarized in Section 12.1 and future perspectives for potential research are given in Section 12.2.

12.1 Key takeaways

12.1.1 Pneumatic conveying

Chapter 2 provided a comprehensive literature review of the present state of research on U_{pu} .

In Chapter 3, the impact of the width of lognormal particle size distributions (PSDs) and particle sphericity (ϕ) on U_{pu} was determined by the modified weight loss method. Two observations were revealed: (i) as PSD width increases, U_{pu} surprisingly exhibits a non-monotonic behavior (namely, decreases then increases), and (ii) the lower the particle sphericity (ϕ) is, the greater the extent of the non-monotonic behavior becomes. The

discrepancy between the U_{pu} values of the experimental data here and values predicted by available correlations underscores the non-negligible impact of PSD width and particle sphericity, which thereby warrants more understanding and the incorporation of such effects to improve the predictive capability of gas–solid pneumatic conveying.

In Chapter 4, the impacts of particle diameter (d_p), particle density (ρ_p) and particle sphericity (ϕ) on the minimum pickup velocity (U_{pu}) of binary mixtures of Geldart group B particles were investigated. Three types of binary mixtures were tested, whereby the two constituents vary in (i) only d_p (i.e., binary-size), (ii) only ρ_p (i.e., binary-density), and (iii) both d_p and ρ_p (i.e., binary-size-density). The key results were: (1) d_p exerts a more dominant influence on U_{pu} than ρ_p ; (2) particle sphericity (ϕ) has a non-negligible impact on U_{pu} ; and (3) a drag and frictional force balance analysis accounting for particle sphericity gives reasonable predictions of U_{pu} , which in turn provides a mechanistic insight on U_{pu} .

In Chapter 5, the U_{pu} of nanoparticles was reported for the first time. U_{pu} , of six nanoparticle species of different materials (i.e., silicon dioxide (SiO_2), aluminum oxide (Al_2O_3) and titanium dioxide (TiO_2)) and surfaces (i.e., apolar and polar) were determined by the weight loss method. Results show that (1) due to relative lack of hydrogen bonding, apolar nanoparticles have higher mass loss values at the same velocities, mass loss curves with accentuated S-shaped profiles, and lower U_{pu} values; (2) among the three species, SiO_2 , which has the lowest Hamaker coefficient, exhibited the greatest discrepancy between apolar and polar surfaces with respect to both mass loss curves and U_{pu} values; (3) $U_{mf,polar}/U_{mf,apolar}$ was between 1 – 3.5 times that of $U_{pu,polar}/U_{pu,apolar}$ due to greater extents of hydrogen bonding associated with U_{mf} ; (4) U_{pu} values are at least an order-of-magnitude lower than that expected from the well-acknowledged U_{pu} correlation (Kalman et al., 2005) due to agglomeration; (5) although nanoparticles should be categorized as Zone III (Kalman et al., 2005) (or Geldart

Group C (Geldart, 1973)), the nanoparticles, and primary and complex agglomerates agree more with the Zone I (or Geldart Group B) correlation.

Next, in Chapter 6, to bridge the gap between the nano-scale and the micro-scale, The U_{pu} values of nine alumina particles with particle diameters (d_p) ranging from 5 to 110,000 nm were determined using the weight loss method, then compared against the established pickup Zones (analogous to the Geldart Groups). Results indicated that: (1) U_{pu} varied non-monotonically with increasing d_p , thus revealing the missing link between the nano- and micro-scales; (2) the intermediate particle diameters surprisingly did not agree with any pickup Zone; (3) Zone III (analogous to Geldart Group C) is inadequate for all the nano-scale particles, so new boundaries and a new Zone are proposed.

Collectively, these studies contributed towards furthering the physical understanding of U_{pu} , not only on the micro-scale, but also the previously uninvestigated nano-scale.

12.1.2 Fluidization

In Chapter 7, a comprehensive comparison of the U_{mf} values predicted by the correlations available in literature was provided, which were classified into four types depending on the form of the equation and applied to more popular Geldart Groups A, B and D particles. Furthermore, a comparison of the correlations containing either voidage at minimum fluidization (ε_{mf}) and/or particle sphericity (ϕ) was carried out to investigate the sensitivity to these difficult-to-measure parameters. The following observations are worth highlighting: (i) discrepancies among U_{mf} predictions are presumably attributed to the empirical data-fitting based on limited experimental datasets rather than physical understanding; (ii) the correlations involving an empirical coefficient as an exponent exhibit greater discrepancies (up to 6 orders of magnitude) in U_{mf} predictions than those without; (iii) predictions for Geldart Group A particles generally displayed greater discrepancies across categories, due in part to a

lack of understanding of cohesive forces associated with Group A particles; and (iv) correlations involving voidage (ε_{mf}) and sphericity (ϕ) exhibit more unphysical trends than those without, presumably due to a limited range of ε_{mf} and ϕ experimentally assessed, hence the inclusion of these two parameters increased the errors associated with these correlations.

Chapter 8 presented an up-to-date literature review of the studies on CFBs in context of pressure signal analysis and the core-annulus phenomenon.

The two studies on CFBs in this thesis covered in Chapters 9 and 10 looked into the energy of pressure signals and upflow annulus behavior of Geldart Group B particles in a CFB riser respectively. In the first study (Chapter 9), the effects of material property, operating condition, and riser axial position on the differential pressure (ΔP) signals were evaluated by comparing the energy contained in the wavelet-decomposed signals (E_{Dj}). Results indicate the following: (i) whereas the averaged ΔP values and power spectra of the signals were largely invariant, E_{Dj} values tended to be sensitive to material property and operating conditions throughout the riser; (ii) under conditions of lower solid loading, higher E_{Dj} values were obtained and E_{Dj} was more sensitive to material properties; (iii) among monodisperse particles, the larger and denser particles tended to be most sensitive to operating conditions; (iv) among the five particle systems, monodisperse small glass and monodisperse large glass systems gave, respectively, the lowest and highest E_{Dj} values; (v) particle density exerts a more dominant influence than particle diameter on E_{Dj} ; and (vi) the effect of axial position on E_{Dj} was secondary compared to that of operating condition and material property. In the second study (Chapter 10), solids flow direction near the wall of a CFB riser was studied for three monodisperse, two binary mixtures and one continuous particle size distribution (PSD) of Geldart Group B particles. Results showed that (i) for monodisperse systems, particle diameter was more dominant in dictating upflow annulus behavior; (ii) the binary mixtures exhibited largely upflow annulus behavior, presumably due to the dominant influence of the larger

constituent particles in dictating upflow annulus behavior; (iii) for the continuous particle size distribution (PSD), the impact of this type of polydispersity was more pronounced at lower solid loading conditions; and (iv) the applicability of the available regime maps, which were largely developed based on Geldart Group A particles, was found to be limited for the Geldart Group B and non-monodisperse particle systems investigated in this study.

12.1.3 Reactor modeling

The reactor modeling study presented in Chapter 11 endeavored to apply a two-phase heterogeneous model to a carbon-dioxide (CO_2) methanation bubbling fluidized bed reactor and assess the effects of varying operating conditions and bed properties on the generation of Synthetic Natural Gas (CH_4). The modelling results indicate that the outlet CH_4 concentration varies non-monotonically, namely increases, then decreases with temperature; decreases with increasing inlet superficial gas velocity; remains almost constant, then decreases with increasing particle diameter; remains unaffected by particle density; and remains almost constant, then decreases with increasing particle sphericity and bed voidage at minimum fluidization. Also, expectedly, CH_4 concentration increases with increasing pressure and exhibits a non-monotonic relationship with $\text{H}_2\text{:CO}_2$ inlet ratio, namely increases then decreases peaking at the stoichiometric ratio.

12.2 Future perspectives

Chapter 3 revealed the impact of PSD width and particle sphericity on the minimum pickup velocity, U_{pu} . As mentioned in section 3.1.5, a method to assess the effects of segregation and examine the size range of the particles entrained from the sample would be to measure the PSD of the residue left behind. Unfortunately, to perform this task on such a small sample was not feasible in the present laboratory. Therefore, an interesting future undertaking would be to determine the PSD of the remnants by testing a larger sample or employing more

sophisticated equipment. This study also highlighted the need to incorporate the effects of PSD widths in the development of such correlations as in the Three-Zone Model.

Chapter 4 provided mechanistic insights into U_{pu} through a drag and frictional force balance analysis by incorporating sphericity and bidispersity into the model. The Brazil nut phenomenon (Rosato et al., 1987), whereby the larger particles preferentially segregate to the top, could have been at play when the more dominant influence on U_{pu} of a binary-size-density (different particle density and diameter for both components) mixture was exerted by the larger constituent. However, the same was not strictly true for the denser constituent (Mobius et al., 2001). This interplay between the larger, less dense component and the smaller but denser component was not fully understood. Future studies could be targeted towards gaining more mechanistic insights into the pickup of binary-size-density mixtures.

Chapter 5 represents the first systematic study of the pneumatic conveying of nanoparticles, which sheds light on the inadequacy of the current U_{pu} correlations mostly based on micron-sized particles and the non-negligible effect of hydrogen bonding in inter-particle cohesion. In particular, apolar SiO_2 nanoparticles exhibited the largest deviations from the correlations for both primary and complex agglomerates. SiO_2 also gave the largest differential between the polar and apolar species in the mass loss curves, U_{pu} values, and also U_{mf} values (Tahmasebpour et al., 2013). Therefore, SiO_2 makes an interesting future case study on the interplay of hydrogen bonding and Hamaker attractions in nanoparticle agglomeration behavior.

Chapter 6 attempted to bridge the gap between the micro-scale and the nano-scale in terms of U_{pu} . Due to the inadequacy of the Three-Zone Model (Kalman et al., 2005) in correctly predicting the U_{pu} of nano-scale particles, modification to the model were proposed. This indicates that the U_{pu} of nano-scale particles deviates from that expected from understanding based on micro-scale particles. This underscores the need for bridging the gap between the two

scales. More datapoints for the proposed modifications to the Three-Zone model would be beneficial. Future studies could be directed at comparing U_{pu} values of a variety of nanoparticles against the modified Model and improve the proposed Zone IV correlation.

A concern common to both the nanoparticle U_{pu} studies (Chapters 5 and 6) was the failure to experimentally determine the PSD of nanoparticles. This meant that the d_p values given by the vendors had to be relied upon. Hence, a corollary investigation could be to test the U_{pu} of the same species purchased from different vendors and compare results. Also, more effective dispersion techniques and/or better equipment need to be used to measure PSD.

Two problems were common to all U_{pu} studies (Chapters 3-6). Firstly, current experimental methods to determine U_{pu} have their limitations like reliance on visual observation (Cabrejos and Klinzing, 1992, 1994), inability to determine U_{pu} accurately for fine particles for the (Kalman et al., 2005), reliance on linear extrapolation of weight loss curve for the ‘decreasing velocity weight loss’ method (Goy et al. (2011); (Tay et al., 2012) and Chapters 5 and 6), and slight overprediction of U_{pu} , for the ‘increasing velocity weight loss’ method (Chapters 3 and 4). Future studies could investigate into these drawbacks and try to develop a novel method to experimentally determine U_{pu} , devoid of said limitations. Secondly, the resolution and magnification of the high-speed camera used for filming particle rotation (Chapter 3) and nanoparticle agglomerates (Chapter 5) was limited. Regarding the former, with the aid of a high-resolution better magnifying camera, the PSD of entraining particles could be monitored to assess pre-existing segregation in samples, and the rotational speed of the entraining non-spherical particles could be measured with the help of image processing softwares. Regarding the latter, efforts could be directed at gaining more physical insights into nanoparticle agglomerates using high-resolution high-speed imaging and image-processing.

Simulation studies on incipient pneumatic conveying, specifically, U_{pu} are lacking. Future simulation works on U_{pu} could aim at (i) supporting experimental findings, (ii) including a wide range of particle and/or fluid properties such as density, particle shape, particle diameter, fluid viscosity, and test whether trends revealed in experiments hold and (iii) gaining a more mechanistic understanding of the pickup phenomenon.

Chapter 7 contended that the correlations available in literature for predicting U_{mf} are highly empirical and system-specific, which severely limit the utility of each to systems beyond the scope tested. Recommendations for future studies on improving the prediction of U_{mf} include using empirical coefficients with caution, accounting for interparticle interactions, especially for Geldart Group A particles, directing greater attention towards bed voidage and particle sphericity values and accurately reporting particle properties, size distributions and exact column configurations.

Chapter 9 tried to assess the effects of material property, operating condition, and riser axial position on the energy of differential pressure signals in a CFB riser via wavelet decomposition. Collectively, the observations in Chapter 9 provide additional insights into not only the mechanistic relationship between the gas phase and the particles but also information available from the ubiquitous pressure transmitters in fluidized bed systems which can find potential use in practical operations. Regarding the former, the enhanced turbulence caused by larger particles has been observed in simulations (Bolio et al., 1995; Rao et al., 2012) but has been difficult to experimentally investigate. The higher E_{Dj} values of the large glass beads (i.e., the larger, denser monodisperse system) revealed in this work provides experimental evidence of the phenomenon. As for the latter, since the influence of operating conditions and material properties on E_{Dj} values was found to be detectable throughout the riser, unlike the $\Delta P/\Delta h$ values, a change in the hydrodynamics at any height in the riser can be readily monitored through E_{Dj} values, which is beneficial for troubleshooting, optimization, and

process control. As wavelet analysis retains the time stamp of the raw signal, it can be used to recognize changes in the unit in real time. More experimental data and fine tuning (e.g., choice of scale, data acquisition frequency) of such a method is needed before benefits in detecting finer changes like particle-size distribution and/or shape due to attrition can be fully harnessed. Nonetheless, because the ability to fully assess the pressure signals in a CFB riser remains largely untested due in part to the lack of a comprehensive experimental data set, the results of this work are expected to find value in furthering the utility of the omnipresent pressure signals.

Chapter 10 studied the solids flow direction near the wall of a CFB riser for three monodisperse, two binary mixtures and one continuous particle size distribution (PSD) of Geldart Group B particles. Limitations of the presently available regime maps underscored the difference between Geldart Group B and the more widely studied Geldart Group A, and the influence of polydispersity. Future studies could be directed at modifying present regime maps or developing new ones to incorporate Geldart Group B particles through expanding the experimental database and via simulations.

Chapter 11 presented a bubbling fluidized bed CO₂ methanation reactor modeling study. Future work should be directed towards gathering experimental data to validate the model. Further avenues for research could be development and testing of new reactions or kinetics in bubbling fluidized bed reactor models, comparing the performance of existing kinetics, comparing carbon-monoxide (CO) and CO₂ methanation in bubbling fluidized bed reactors, development of detailed and two-dimensional models, changes in design for the fluidized bed (addition of internals like stirrers and fins, tapered fluidized beds).

12.3 Overall conclusion

The research presented in this thesis has thus contributed in providing mechanistic insights into horizontal pneumatic conveying, minimum fluidization velocity, gas-solid interaction in CFB risers and CO₂ methanation in fluidized beds, especially in the context of the impact of operating conditions and particle properties. Knowledge gaps were identified keeping in mind the underlying thread of behavioral differences in and across Geldart Groups (and impliedly Pickup Zones) and an attempt was made to bridge them. Five key conclusions were obtained. Firstly, particle diameter (d_p) played a more dominant role than particle density (ρ_p) in dictating behavioral changes. This was evident in the U_{pu} of binary mixtures where the larger species was more dominant in dictating U_{pu} of binary-size mixtures than the denser species was in dictating that of the binary-density mixtures. Also, for binary-size-density mixtures with the larger species was less dense, d_p dominated U_{pu} trends. The dominance of d_p was consistent in annulus flow trends where it dictated upflow annulus and the secondary role of ρ_p was consistent in the methanation reactor model, where CH₄ concentration was unaffected by it. The only exception was energy from pressure signals where ρ_p dominated the trends. Secondly, particle sphericity (ϕ) was an influential parameter, both in horizontal pneumatic conveying and fluidization. U_{pu} trends of continuous particle size distributions (PSDs) and binary mixtures had more pronounced deviations from linearity for less spherical particles and U_{mf} was sensitive to ϕ as seen from the greater discrepancies obtained for U_{mf} correlations containing ϕ . Thirdly, the impact of polydispersity was clearly visible as seen from three observations: (i) the non-monotonic trends of U_{pu} with respect to the width of the lognormal size distributions, which agreed with segregation trends observed in bubbling fluidized beds (Chew et al., 2010), (ii) U_{pu} of lognormal PSDs not agreeing with the Three-Zone Model, which is developed from monodisperse systems and (iii) different annulus flow trends obtained for monodisperse particles and lognormal PSD at the lower solid loading conditions. Fourthly,

inter-particle cohesive effects impact the behavior of Geldart Groups C and A particles strongly as seen from nanoparticles being picked up as agglomerates (which was consistent with the way they fluidize), the need to propose new Pickup Zones for nanoparticles and the greater discrepancies exhibited by U_{mf} correlations for Geldart Group A predictions. Finally, with respect to the physical gas-solid interactions in CFBs, the impact of operating conditions was cognizable, though secondary, to that of material properties; whereas in the methanation reactor model, the impact of operating conditions was more pronounced than that of material properties.

This research effort was thus directed at a fundamental level, mostly addressing the knowledge gaps in physical interactions between the two phases in gas-solid flow. Further work is hence required before these studies can directly impact industry. However, the findings presented here will aid future research in the following manner. Firstly, the experimental understanding and data gained with respect to the U_{pu} of continuous PSDs, binary mixtures and non-spherical particles will aid in the validation of two-fluid models and help enhance the capabilities of such models to adapt the various closures (kinetic theory, drag laws etc.) to the implications of PSD and sphericity. Secondly, the studies on U_{pu} of nanoparticles have started a new database of its kind and will provide a basis for future experimental and/or modeling studies in this area. Thirdly, the comprehensive review of U_{mf} correlations will serve as a guide for future authors trying to develop more physically-based U_{mf} models in particular, and as a reference to students of bubbling fluidized beds in general. Fourthly, the comprehensive experimental dataset and analysis for interpreting differential pressure signals from a CFB riser (Chapter 9) and the riser annulus flow behavior of monodisperse, binary and continuous PSD (Chapter 9) of Geldart Group B particle systems, along with a series of past studies reporting cluster characteristics (Chew et al., 2012a, b), mass flux (Chew et al., 2011b), species segregation (Chew et al., 2011a), radial solids concentration (Chew et al., 2012c) on the same riser are expected to aid in the development of models to provide a more mechanistic

understanding of riser flow behavior as well as triggering more work on flow regime maps in CFB risers. In particular, since pressure signals from CFB risers were capable of detecting changes in material property and operating condition, such analyses could potentially be beneficial for trouble-shooting, optimization and process control. The insights on the annulus flow behavior of Geldart Group B particles will aid in incorporating changes in theory and design to account for such systems in future test units. Fifthly, the reactor modeling study can be used as a basis for experimental studies and can in turn be validated by them. Finally, all studies have made novel contributions to the overall gas-solid flow research database, which will aid future researchers working on experiments, simulations, theoretical models and prototypes.

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List of Publications

- A. Anantharaman, R. A. Cocco and J. W. Chew (2018). "Evaluation of correlations for minimum fluidization velocity (U_{mf}) in gas-solid fluidization", Powder Technology **323**: 454-485.
- A. Anantharaman, J. R. van Ommen and J. W. Chew (2017). "Minimum pickup velocity: The transition between nano-scale and micro-scale." AIChE Journal **63**(5): 1512-1519.
- A. Anantharaman, A. Issangya, S. B. R. Karri, J. Findlay, C. M. Hrenya, R. A. Cocco and J. W. Chew (2017). "Annulus flow behavior of Geldart Group B particles in a pilot-scale CFB riser." Powder Technology **305**: 816-828.
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- A. Cahyadi, A. Anantharaman, S. Yang, S. B. R. Karri, J. G. Findlay, R. A. Cocco and J. W. Chew (2017). "Review of cluster characteristics in circulating fluidized bed (CFB) risers." Chemical Engineering Science **158**: 70-95.