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**THE IMPACT OF PARTICLE POLYDISPERSITY ON
FLUIDIZATION PHENOMENA**

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SCHOOL OF CHEMICAL AND BIOMEDICAL ENGINEERING

2018

**THE IMPACT OF PARTICLE POLYDISPERSITY ON FLUIDIZATION
PHENOMENA**

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THE IMPACT OF PARTICLE POLYDISPERSITY ON FLUIDIZATION PHENOMENA

SUMMARY

Particles naturally found in polydisperse form, but systematic study on the impact of particle polydispersity in fluidization field is overshadowed by the more common particle properties (e.g., diameter, density). Recent development in computational fluid dynamics (CFD) codes and computing capabilities enable detailed study of inter-species interactions in a complex and chaotic particle-fluid flow, hence advancing mechanistic understanding of fluidization phenomena. The impact of particle polydispersity (i.e., monodisperse, binary, ternary, and continuous PSD mixtures) on column hydrodynamics (i.e., local particle and fluid velocities, mixing/segregation, inter-species interactions), fluidization phenomena (i.e., entrainment flux, TDH, cluster characteristics), and system performance (i.e., energy consumption, membrane fouling mitigation in Anaerobic Fluidized Bed Membrane Bio-Reactor/AnFMBR) are studied. Analysis of entrainment and TDH values of monodisperse and polydisperse mixtures (Chapters 2 – 4) showed (i) polydispersity effects are poorly incorporated into fluidization correlation, while the use of empirical constants are inadequate to represent such impact, as indicated by huge discrepancies among prediction of correlations intended for similar system; (ii) physical insights into the relation between particle polydispersity and inter-species interactions, i.e., cohesive agglomeration, cluster formation, and inter-species collisions, are guaranteed. Detailed investigation into cluster characteristics of polydisperse particles in CFB riser revealed (i) lower cluster break-up of monodisperse particle relative to polydisperse particles for complete/partial mixing conditions, because of the higher inter-species momentum transfer for multi-

species mixtures; (ii) under segregated condition, polydisperse particles also hinders cluster formation (i.e., bidisperse < continuous PSD) because only smaller constituents form clusters at centre column, while larger particles concentrated near wall area destroys smaller constituent's clusters as they travel radially. Subsequently, fluidization of mono-, bi-, and tri-disperse GAC to induce particle-membrane scouring in AnFMBR are evaluated (Chapters 5 – 7). CFD and experimental results found (i) GAC mean diameter increases particle-membrane scouring (or decreases membrane fouling) through higher superficial liquid velocity required to scour entire membrane height, which indicates the trade-off between fouling mitigation and energy usage; (ii) for fully segregated condition (i.e., high superficial liquid velocity or power input), number of particle constituents (i.e., tridisperse > bidisperse > monodisperse) and size differences among constituents (i.e., larger d_{p2}/d_{p1} and σ/d_{sm} for bidisperse and continuous PSD, accordingly) decrease particle-membrane momentum transfer, because small constituents at upper reactor obtained less energy due to high pressure drop at lower reactor consist of highly-concentrated larger GACs; (iii) the opposite findings of the (ii) conclusion found in partial mixing condition (i.e., low superficial liquid velocity), presumably due to the more dominant inter-species collisions.

Keywords: polydispersity, segregation, hydrodynamics, two-fluid model, gas-solid fluidization, anaerobic fluidized bed membrane bioreactor

CHAPTER 1: INTRODUCTION

1.1. Background

Particle polydispersity affects hydrodynamics of fluidized bed, such as system pressure drop [1, 2], pressure fluctuation [3], minimum fluidization velocity [4-6], bubble characteristics (i.e., minimum bubbling velocity, bubble size, and the amount of particles inside bubbles) [1, 4, 7], particle segregation [8, 9], the amount of particle clusters [1, 10], entrainment rate [2-6, 11], and interphase mass transfer [12]. The impact of polydispersity is often associated with the presence of fine particles ($d_{pi} < 40\mu\text{m}$) [11] in relation with their impact on inter-particle force or inter-particle momentum transfer [2, 13-17]. In view of the complex and chaotic nature of particle-fluid flow, the impact of particle polydispersity on fluidization behavior was accounted in the embodiment of *ad hoc* constants during data fitting process of fluidization correlations. Unsurprisingly, available semi-empirical equations have very limited applicability, while design, optimization, and scale-up of fluidized bed systems remain as empirical science [18, 19]. Furthermore, contradictory findings on the impact of particle polydispersity towards fluidization behaviors are common due to the lack of systematic study on the impact of particle polydispersity towards fluidization phenomena [5, 20-27]. The need of a more mechanistic understanding on fluidization phenomena is of importance to meet industry demand of a more reliable future-fluidized bed technology, as well as to optimize current fluidized bed technology.

1.1.1 Fluidization

Fluidization is a physical process which particles are suspended by constant up-flowing fluid flow, either liquid or gas [28]. Suspended particles increase inter-phase

interactions compared to fixed bed process, resulting in higher heat and mass transfer, and near isothermal condition within fluidized bed. Since its first application in industry in 1920s, fluidized bed systems have been used widely in industries as gasifier, combustor, dryer, and reactor (e.g., chemical, pharmaceutical, petrochemical, metallurgical, electronics, fast moving consumer goods (FMCG), and wastewater industries) [29].

Fluidization regime or flow behavior changes accordingly with the magnitude of superficial fluid velocity (i.e., fluid velocity measured from the assumption that particles presence which reduce cross-sectional flow area is negligible). Fig. 1.1 depicts relationship between superficial fluid velocity, pressure drop across fluidized bed, and fluidization regime. It is worth noting that fluidization regime is also affected by particle properties (e.g., Geldart classification, as explained in the next section) and fluid properties (e.g., liquid-solid or gas-liquid-solid fluidization).

Pressure drop across particle bed and bed height increase linearly with superficial fluid velocity before fixed bed is fully suspended as fluidized bed. Fluidization happens as pressure drop across bed is equal to bed weight per cross-sectional area of the column (W/A_c). At superficial fluid velocity beyond minimum fluidization velocity (U_{mf}), dense fluidization regime (i.e., particulate and bubbling regimes) occur as pressure drop across column is constant and particle entrainment (i.e., particles carried by fluid flow out of the column) is negligible. Bubbling fluidization (i.e., at superficial fluid velocity between minimum bubbling velocity (U_{mb}), and U_c) is characterized with the presence and continuous growth and coalescence of bubbles (gas) which promotes particle-gas mixing. Higher superficial fluid velocity results in dilute fluidization regime (i.e., turbulent fluidization beyond superficial fluid velocity of U_k , and fast fluidization and pneumatic conveying beyond transport velocity (U_{tr})) in which

particle-fluid demarcation line started to diminish and entrainment flux is non-negligible. It is worth mentioning that liquid-solid fluidization is solely considered as particulate fluidization before particle entrainment is dominant.

Fig. 1.1. Relationship between superficial fluid velocity, pressure drop, and fluidization regime [6].

In this thesis, Chapters 2 – 4 focus on dense fluidization regime, namely bubbling and turbulent regimes in which complex hydrodynamics instabilities (e.g., clusters or bubbles) are more pronounced, while Chapters 5 – 7 evaluate particulate fluidization in solid-liquid fluidized bed.

1.1.2. Geldart Groups classification

Geldart classification (i.e., Group A, B, C, and D) are developed to predict gas-solid fluidization behaviour of monodisperse particles at ambient conditions based on particle diameter and density (see Fig. 1.2) [30]. Group A particles characterized as relatively small particles with low particle density ($\leq 1400 \text{ kg/m}^3$) which undergo particulate fluidization, followed by bubbling fluidization, as superficial gas velocity increases beyond minimum fluidization velocity. Geldart Group B particles (i.e., most of particles with diameter between $40 - 500 \text{ }\mu\text{m}$ and density of $1400 - 4000 \text{ kg/m}^3$) indicated by the absence of particulate fluidization above minimum fluidization velocity (i.e., $U_{mb} = U_{mf}$). Fine particles in Geldart Group C exhibit strong cohesive force compared to hydrodynamics force which prevent homogeneous particle fluidization (i.e., channeling flow). Large particles of Geldart Group D distinguished from other type of particles by their spouting-type of fluidization. Table 1.1 provided characteristics flow behaviour of different Geldart Group particles.

Most of the particles in gas-solid fluidization industry classified under Geldart Group A and B particles owing to their superior gas-solid mixing (see Table 1.1), which is advantageous to promote inter-phase mass and heat transfer, reaction kinetics, and homogeneous temperature distribution along fluidized bed system [30]. Subsequently, Chapters 2 – 4 in current thesis evaluated fluidization phenomena of monodisperse and polydisperse particles classified as Geldart Group A and B particles based on their mean-particle diameter. It is worth noting that the flow behaviour of particles used in liquid-solid fluidization (Chapters 5 – 7) could not be evaluated according to Geldart classification.

Fig. 1.2. Geldart Group classification [30].

Table 1.1. Characteristics of Geldart Group A, B, C, and D particles [30].

	Group C	Group A	Group B	Group D
Example	Flour, cement	FCC catalyst	Sand-like particles	Biomass particles
Flow behaviour	Difficult to be fluidized due to cohesive forces	Particulate, followed by bubbling fluidization	Bubbling fluidization	Spouting flow even in deep bed
Expansion bed height	Small (channeling)	Large	Moderate	Small (spouting)
Particle mixing	Very small	Large	Moderate	Small
Gas back-mixing	Very small	Large	Moderate	Small

1.1.3. Particle polydispersity

Monodisperse and polydisperse systems are characterized with negligible particle size distribution (i.e., $\text{PSD} \leq 10\%$), and wider size and/or density variations within particle mixtures, respectively [11]. PSD width in fluidization experiment could be measured offline through particle sampling followed by sieving analysis [31]. Advanced non-invasive methods based on radioactive particle tracking, or diffraction patterns of the laser beam upon direct contact on particle surface to reconstruct particle's projection images enable online and localized PSD measurement during particle fluidization [32]. Such methods results in 2D or 3D particle's projection images, with the former may overestimate the PSD of elongated particle mixtures [33].

Optical-based PSD measurement and particle sieving methods produce results in number-based and mass-based frequency distribution, respectively [34]. There are three PSD functions can be used to express polydispersity of continuous PSD systems, namely normal, lognormal, and Rosin-Rammler PSDs. Lognormal PSD is suitable for crushed and grinded particles (e.g., GAC), while Rosin-Rammler PSD commonly used for severely crushed particles with extremely skewed normal PSD graph [4]. Subsequently, all studies in current thesis uses lognormal PSD function as explained in detail in each chapter. Lognormal distribution offers two advantages compared to normal PSD, i.e., easier conversion of various equivalent diameter (e.g., number-based or mass-based) into statistical diameter, and single magnitude of mean, median, and mode eliminates possible error due to the use of different statistical characteristics to represent the central tendency of particle size [1].

1.2. Motivation and Objectives

1.2.1. Overall objectives

Particles in nature and industrial processes exist in polydisperse form (i.e., mixture of various size and/or density, such as binary or continuous particle size distributions (PSDs) system). Nevertheless, most of the studies on particle-fluid flow, such as fluidization, used monodisperse particles (i.e., homogeneous size and density) or expressed size polydispersity merely using mean particle diameter of the mixture. Such simplification to quantify particle polydispersity does not improve recent effort to develop more reliable fluidization equations, nor physical understanding of fluidization phenomena based on the first-principle. Subsequently, the first three chapters of this thesis (Chapters 2 – 4) aim to elucidate the extent of particle polydispersity impact on key fluidization phenomena, namely entrainment, TDH, and particle clustering, which directly related to the design, operation, scale-up, and optimization of fluidized bed systems. Following up the finding on the prominent impact of particle polydispersity on fluidization phenomena, the last three chapters of this thesis (Chapters 5 – 7) aim to provide detailed study, based on Two Fluid Model (TFM) simulation, on the impact of particle polydispersity on system hydrodynamics and particle scouring efficiency to mitigate membrane fouling in Anaerobic Fluidized Bed Membrane Bioreactor (AnFMBR).

1.2.2. Particle polydispersity effects on current predictive capability of fluidization phenomena (entrainment flux, TDH, particle clustering)

Prediction of entrainment flux (Chapter 2) and TDH (Chapter 3) are crucial in the development of fluidized bed systems, particularly to minimize particle bed loss, while optimizing cyclone design and operation aimed to re-capture particles within

entrainment flow. Abundant amount of design correlations to predict entrainment flux and TDH are available in the literature due to the limited applicability of such correlations within certain operating conditions and/or particle types (as expressed in Geldart Group classification). Current studies aim (1) to evaluate the reliability of entrainment and TDH correlations to predict the performance of polydisperse systems, which is crucial in real application but rarely studied during the development of such correlations; (2) to elucidate the impact of particle polydispersity on entrainment and TDH phenomena. Among other fluidization phenomena, the lack of mechanistic understanding on dynamic particle clustering hinders current effort to develop first-principle based fluidization equation. In order to compliment other studies which commonly evaluate the impact of particle size and density on cluster characteristics, Chapter 4 reviewed the impact of particle polydispersity on particle cluster characteristics (i.e., cluster size, duration/growth, frequency/formation, and appearance probability).

1.2.3. CFD study on the impact of particle polydispersity on fluidized bed system performance.

Particle-fluid flow, such as fluidization, is complicated phenomenon owing to the dynamic interactions among particles and between particle-fluid. As such, fluidization is commonly perceived as an empirical science [2, 3], which rely on empirical coefficients obtained from experiment to derive design equations. Expectedly, the lack of mechanistic understanding on fluidization phenomena has prompted constant issues on the scale-up and non-optimum operation of fluidization systems. The rapid development of CFD codes and computing capabilities enable more rigorous evaluation of fluidization phenomena which is hard, if not impossible, to be evaluated using

existing experimental measurement equipment. Such variables which compliment experimental results include local particle and fluid velocities, local particle collisions or interaction forces, etc. Furthermore, computational study allows more systematic study by isolating certain design variables, which are not practical to be done in experiment, but may provide physical insight into fluidization behavior. Subsequently, Two-Fluid Model simulation on the fluidization behavior of mono-, bi-, and tri-disperse granular activated carbon (GAC) inside Anaerobic Fluidized Bed Membrane Bioreactor (AnFMBR) are conducted. Detailed hydrodynamics data indicate efficiency (i.e., energy consumption), and effectiveness of particle-membrane scouring qualitatively (i.e., scouring distribution) and quantitatively (i.e., scouring magnitude). Such indicators are linked to the results of fouling experiment in order to evaluate the relation between membrane fouling mitigation (i.e., trans-membrane pressure (TMP)) and particle-membrane scouring.

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CHAPTER 2: PREDICTION OF PARTICLE ENTRAINMENT FLUX FOR MONODISPERSE AND POLYDISPERSE SYSTEMS

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Abstract

Despite the abundance of entrainment correlations in gas-solid fluidization made available in the past few decades, the discrepancy between empirical prediction and experimental data has been noted to be up to a hundredfold in some cases. Hence, a comprehensive review of the available correlations is warranted, with the goal of extracting the underlying physics giving rise to the disparities, and thereby providing insights towards an enhanced understanding of entrainment. This review addresses a comprehensive spectrum of particle systems, ranging from monodisperse to binary to continuous particle size distribution (PSD) systems for the more predominant Geldart Groups A and B classifications. Three key observations are highlighted. First, comparisons of the predicted entrainment values among available correlations reveal discrepancies spanning several orders of magnitude, with the maximum being 20 orders of magnitude, evidencing that available empirical correlations do not extrapolate well beyond the scope of tested parameters. Second, unphysical phenomena predicted include qualitative discordance on the shapes of the elutriated PSDs, the most notable

of which is the anomaly whereby larger particles are preferentially elutriated. Third, whether a particular correlation was developed for a specific Geldart Group or for monodisperse and/or polydisperse systems seems inconsequential to giving better predictions even when appropriately applied to the particular subset. These observations underscore the need for more physically-based models to enable a more accurate prediction of entrainment and elutriation phenomena. In particular, the lack of understanding of inter-particle effects, namely cohesive agglomeration of smaller particles, dynamic clustering due to hydrodynamic instabilities, and inter-species collisional momentum transfer seem largely culpable for the huge discrepancies in the predicted K_{∞} values.

Keywords: binary; continuous particle size distribution; elutriation; entrainment; gas-solid fluidization; Geldart Groups A and B; monodisperse

2.1. Introduction

One of the first correlations for entrainment was published in 1955 [1]. Since then, an abundance of entrainment correlations has been made available [2-8]. Unfortunately, due to the use of empirical constants in the absence of a more fundamental understanding of such processes, the discrepancy between empirical prediction and experimental data is up to a hundredfold in some cases [4, 7]. Good predictive capability of entrainment values is critical for the technical and economic success of a fluidized bed application, because the optimal design of gas-solid separators like cyclones and filters [4, 8] to minimize bed material loss hinges on such a priori estimates. Hence, a comprehensive review of the available correlations is warranted, with the goal of extracting the underlying physics giving rise to the disparities, and

thereby providing insights towards enhanced understanding of entrainment and elutriation. It is worthwhile to note three items regarding the scope of this review: (i) new experimental data is not presented, since one of the key failings of empirical entrainment correlations appears to be that the underlying experimental data is only relevant to the narrow set of conditions it was taken at and may not be appropriate for extrapolation, (ii) recommendation on which of the correlation is most relevant is beyond the scope of this review, since such clarity is not possible without more research work into better physical understanding and description of the entrainment phenomenon, and (iii) recommending rules-of-thumb on reactor design is not part of this review.

The difference in definition of the terms entrainment and elutriation needs clarification. Although both generally refer to the flow of solids out of fluidized beds, they cannot be used interchangeably especially for polydisperse (i.e., the presence of a range of particle sizes and/or particle density) systems. On one hand, entrainment refers to the overall flux of solids carried out of the fluidized bed by gas; on the other hand, elutriation refers to the classifying effect of fluidized bed entrainment, specifically characterizing the selective removal of particles of individual sizes from the fluidized bed [4, 8]. In other words, while entrainment flux is equal to elutriation flux when only a single species is present as is the case for monodisperse systems, entrainment flux is greater than the elutriation flux for each species in the presence of multiple species for polydisperse systems.

Physically, entrainment in the freeboard is widely acknowledged as being initiated by the role of bubbles in ejecting particles upwards [8-10], although different schools of thoughts exist on whether particles ejection stems from the bubble nose or bubble wake [8]. The different origins have been reported to cause discrepancy of up to one order of magnitude in the entrainment flux at the bed surface [8], due primarily to

differences in (i) particle size distribution above the bed surface, and (ii) particle dispersion in the freeboard [11, 12]. Regarding (i), the bubble nose is observed to preferentially eject fine particles, whereas the bubble wake ejects both fine and coarse particles. With respect to (ii), the bubble nose aids dispersion of particles after being ejection from the bed surface, whereas the bubble wake ejects particle clusters into the freeboard. As particles travel upwards in the freeboard, some particles will return to the dense bed while others will continue the upward trajectory, thereby resulting in an exponentially decreasing entrainment or particle concentration profile with respect to height. For more details on the entrainment phenomenon, the interested reader is referred to the book chapters [2, 4, 6, 8].

Experimental efforts targeted at quantifying entrainment and elutriation have been pursued, resulting in a broad spectrum of empirical constants rooted in data-fitting and varying emphasis on different system or operating parameters being presented. Modeling efforts have also progressed in parallel. As early as in the 1950s, simplified models developed based on particles behaving independently [10, 13] proved insufficient in explaining observed phenomena like the entrainment of particles with terminal velocities greater than the operating superficial gas velocities ($U_t > U_g$) [14, 15], and decreasing entrainment and solids concentration with increasing height. To date, due to the complexity of gas-solid flows, particularly with respect to inter-particle cohesion and hydrodynamic clustering effects, models continue to rely on assumption-based simplified physical descriptions. Experiments and models are complementary tools: while experimental results provide data for validating models, models are conversely necessary to extract the physics underlying the observed flow phenomena [16]. Accordingly, the generation of an understanding of the physics underlying

available empirical entrainment correlations is necessary in order to validate and improve models; generating such an understanding is an overall aim of this work.

In the current review, K_{∞} refers to the total entrainment flux above transport disengagement height (TDH), while $K_{i\infty}$ refers to the elutriation flux of individual species i above TDH. Specifically, K_{∞} and $K_{i\infty}$ are related by the equation $K_{\infty} = \sum x_i K_{i\infty}$, where x_i is the mass fraction of species i in the dense bed. Regarding TDH, it is defined as the height of a fluidized bed column above the dense bed and splash zone beyond which the solids concentration becomes relatively constant [2, 4, 6, 8]. Notably, preliminary comparisons show that predictions of TDH by various correlations are about an order of magnitude apart [17-20], hence correlations which incorporate TDH as a parameter [21-23] are left out of the review to avoid potential errors in the corresponding K_{∞} , which is defined for entrainment above TDH. An effort was made to collect all available entrainment correlations available in literature, as compiled in Table 2.1 in chronological order. It is worth mentioning that correlations without complete validity range of particle properties (i.e., density and/or particle range) are assumed to be valid throughout Geldart Group classification of the particles used in the original experiment. Note that the terms ‘single species’ and ‘multiple species’ are used in the table instead of ‘monodisperse’ and ‘polydisperse’, respectively, because no indication of the actual PSD of the initial bed was given.

This review encompasses a wide spectrum of particle systems, including monodisperse, binary, and continuous particle size distributions (PSDs) for the more ubiquitous Geldart Groups A and B particles [24]. The superficial gas velocity (U_g) values evaluated spans from a ratio of superficial gas velocity to terminal velocity (U_g/U_t) of 1.01 (incipient entrainment) to 2 (approximately critical velocity, U_{se} , defined as the

point where the solids begin to be entrained significantly as the system begins to transition into the fast fluidization regime [25, 26]).

On the effect of polydispersity, several authors qualitatively attributed the effect of fine particles ($d_{pi} < 40\mu\text{m}$ [27]) on K_∞ to inter-particle cohesive forces or the clustering phenomenon [28-33]. Notably, K_∞ was found to be reduced by up to two orders of magnitude in the presence of 30 mass % of 10 μm particles [34], although the effect of fine particles on K_∞ is reduced as gas velocity increases [35]. It is also generally accepted that the presence of the particles with different size and/or density will affect K_∞ due to inter-particle momentum transfer, which allows particles whose U_t is greater than U_g to be entrained [14, 15], but such a description is not available yet. Regrettably, despite the well acknowledged impact of polydispersity on entrainment, only a few authors specified the composition of fine particles [1, 14, 23, 28, 36-39] and none specified the PSD of the initial bed used to develop the entrainment correlations. Inclusion of the effects of polydispersity is generally attempted through the *ad hoc* inclusion of terms to enhance data-fitting in the development of entrainment correlations, which leads to huge discrepancies among the K_∞ values predicted, as will be illustrated in this review. An in-depth study on the impact of polydispersity is necessary because it is prevalent; however, a predictive understanding of fluidized-bed processes with polydispersity remains elusive [40-43].

Another four contributors to huge discrepancies in K_∞ values is worth noting. The first lies in the height at which the experimental data was collected at, since discrepancies in predicted TDH values of up to a few orders of magnitude have also been reported [44]. In particular, because the K_∞ value and relative composition of the various particle sizes changes with height [21], experimental data collected at different heights will lead to the development of different correlations. Second, temperature [23,

37, 45-47] and pressure [13] effects have non-negligible impact on entrainment and elutriation too, due to the corresponding effect on gas viscosity and density; unfortunately, such parameters are largely not incorporated into the correlations. With regards to temperature, K_{∞} has been found to trend non-monotonically with respect to temperature; specifically, K_{∞} appeared to decrease as temperature increases at lower temperatures, then increases with temperature in the range of 293-873 K [48]. Another author has instead found that the effect of temperature on K_{∞} can be ignored in the range of 300-445 K [37]. Such inconsistencies are presumably system-dependent and needs to be reconciled through more fundamental understanding, otherwise good predictive capability of K_{∞} is not achievable. Third, relative humidity (RH) [23, 31, 49] also plays a role in entrainment and elutriation, despite the fact that the RH value is generally not reported. It is acknowledged that RH has to be maintained above a certain threshold (typically, 40 %) to minimize electrostatic charges between particles [50-53], although too high an RH decreases K_{∞} due to the increase in inter-particle forces due to the liquid bridging effect [54, 55]. Briens et al. [56] showed that neutralizing electrostatic charges increases K_{∞} by up to two orders of magnitude, and that electrostatic charge is more dominant for smaller particles and at lower temperatures. Also, even though electrostatics does not promote clustering appreciably [54], K_{∞} was found to be reduced due to the increased interactions between the column wall and the particles [54]. Forth, the experimental setup affects the entrainment data and hence the correlations developed obtained too. Entrance and exit effects are well-documented, so the fact that some correlations were developed employing a circulating system [13, 22, 23, 35-37, 39, 45, 47, 49, 57-60], and some not [1, 27, 28, 31, 38, 39, 57, 61-63] is expected to cause discrepancies in the correlations. Also, the effect of the column geometry (e.g., rectangular versus circular cross-section) and column diameter on K_{∞} is also important,

especially when a two-dimensional column is used [13] wherein wall friction cannot be ignored [64]. In summary, the use of empirical constants in the existing correlations to data-fit experimental results undoubtedly gives excellent predictive capability for each specific system, but applicability beyond is limited without a fundamental understanding and description of the effect of each variable.

Remarkably, discrepancies of up to 20 orders of magnitude exists among the predicted K_{∞} values of available correlations, indicating that empirical constants and data-fitting impedes the use of the correlations beyond the breadth of specific particles and apparatus. In addition, the unphysical phenomenon whereby larger particles are preferentially elutriated is predicted by a handful of correlations. Also, whether a particular correlation was developed for a specific Geldart Group or for monodisperse and/or polydisperse systems seems inconsequential, since outlying K_{∞} values do not necessarily imply application to a different Geldart group than was developed for. These observations highlight the need for more physically-based models to enable a more accurate prediction of the elutriation and entrainment phenomena.

Table 2.1. Entrainment correlations.

Year	Authors	Correlation	Geldart Group	ρ_p (kg/m ³)	d_p (μm) (% wt of fines)	U_g (m/s)	D_c (m)	H_c (H_s) (m)
1955	Yagi and Aochi [1]	$K_{i\infty} = \frac{\mu_g (U_g - U_{ti})^2 (0.0015 Re_{ti}^{0.6} + 0.01 Re_{ti}^{1.2})}{g d_{pi}^2}$	A, B; multiple species		85-500; 310- 1640 (5-20)	0.92- 1.62	0.051- 0.071	(0.031 -0.12)
1958	Zenz and Weil [13]	$K_{i\infty} = 1.26 \times 10^7 \rho_g U_g \left(\frac{U_g^2}{g d_{pi} \rho_p^2} \right)^{1.88} \text{ for } \frac{U_g^2}{g d_{pi} \rho_p^2} < 3 \times 10^{-4}$ $K_{i\infty} = 4.31 \times 10^4 \rho_g U_g \left(\frac{U_g^2}{g d_{pi} \rho_p^2} \right)^{1.18} \text{ for } \frac{U_g^2}{g d_{pi} \rho_p^2} > 3 \times 10^{-4}$	A; multiple species		10-150	0.3- 0.72 ($P=7.8$ atma)	2-D column; 0.051x0. 61	4.3

1960	Wen and Hashinger [28]	$K_{i\infty} = 1.52 \times 10^{-5} \rho_g \frac{(U_g - U_{ti})^2}{g^{0.5} d_{pi}^{0.5}} Re_{ti}^{0.725} \left(\frac{\rho_p - \rho_g}{\rho_g} \right)^{1.15}$	A, B; multiple species	2499; 1298	71-279; 97-714 (6-100)	0.22- 1.32	0.051- 0.102	1.83- 2.08
1972	Tanaka et al. [57]	$K_{i\infty} = 4.6 \times 10^{-2} \rho_g \frac{(U_g - U_{ti})^2}{g^{0.5} d_{pi}^{0.5}} Re_{ti}^{0.3} \left(\frac{\rho_p - \rho_g}{\rho_g} \right)^{0.15}$	B; multiple species	1540- 2670	60-2300	1.28- 2.7	0.067	1.8 (0.25)
1974	Merrick and Highley [45]	$K_{i\infty} = \rho_g U_g \left[A + 130 \exp \left(-10.4 \left(\frac{U_{ti}}{U_g} \right)^{0.5} \left(\frac{U_{mfi}}{U_g - U_{mfi}} \right)^{0.25} \right) \right]$	A, B; single species	2400	125- 2000	0.61- 2.44 (T=973 -1173 K)	Square cross- section; 0.91x 0.91;	3.96 (0.61- 1.22)

							0.91x0.4 5	
1979	Geldart et al. [14]	$K_{i\infty} = 23.7\rho_g U_g \exp\left(-5.4 \frac{U_{ti}}{U_g}\right)$	A, B; multiple species	706- 5000	62-219 (5-75)	0.6-3	0.076	3.8 (0.35- 0.45)
1980	Lin et al. [36]	$K_{i\infty} = 9.43 \times 10^{-4} \rho_g U_g \left(\frac{U_g^2}{g d_{pi}}\right)^{1.65}$	C, A, B; multiple species	860	0-125, 125-419 (0.01-1)	0.1-0.3 ($P=1$ - 1.5 atm)	Square cross- section;0 .61x0.61	0.88- 3.52 (0.25)
1981	George and Grace [37]	$K_{i\infty} = 0.728 U_g^{5.2}$ (without column internals)	A, B; multiple species,	2630	30-272 (50)	0.2-1.3 ($T=300$ -445 K)	Rectangu lar cross- section;	3 (0.54)

			column internals				(0.254- 0.432 m)	
1981	Colakyan et al. [65]	$K_{i\infty} = 33 \left(1 - \frac{U_{ti}}{U_g}\right)^2$	B; for $d_p > 100 \mu\text{m}$					
1982	Wen and Chen [66]	$K_{i\infty} = \rho_p(1 - \epsilon_i)(U_g - U_{ti})$ $\epsilon_i = \left(1 + \frac{\lambda(U_g - U_{ti})^2}{2gD_c}\right)^{-\frac{1}{4.7}}$ $\frac{\lambda\rho_p}{d_{pi}^2} \left(\frac{\mu_g}{\rho_g}\right)^{2.5} = \begin{cases} 5.17Re_i^{-1.5}D_c^2 & \text{for } Re_i \leq \frac{2.38}{D_c} \\ 12.3Re_i^{-2.5}D_c & \text{for } Re_i \geq \frac{2.38}{D_c} \end{cases}$	A, B; single species					
1982	Matsen [67]	$K_{i\infty} = 0.0003\rho_p \frac{U_g}{U_{ti}} \quad \text{for } \frac{U_g}{U_{ti}} < 1.29$	Not Specified					

		$K_{i\infty} = 2.84 \times 10^{-5} \rho_p \left(\frac{U_g}{U_{ti}} \right)^{3.41} U_g$ for $\frac{U_g}{U_{ti}} > 1.29$						
1984	Colakyan and Levenspiel [38]	$K_{i\infty} = 0.011 \rho_p \left(1 - \frac{U_{ti}}{U_g} \right)^2$ for $U_{ti} < U_g$	A, B; single species	920- 5900	36-1000 (<10)	0.901- 3.66	Square bed (0.92x0.92; 0.3x0.3; 0.1x0.1)	7.5
1985	Kato et al. [58]	$K_{i\infty} = 2.8 \times 10^{-2} U_{ti}^{2.1} \left(\frac{U_g - U_{ti}}{U_{ti}} \right)^{1.6} \left(\frac{\rho_p - \rho_g}{\rho_g} \right)^{0.54} D_c^{-0.63}$	A, B; single species; column internals	1440, 2600	0-500	0.4-1.5	Square bed (0.25x0.25; 0.15x0.15)	2

1987	Kato et al. [59]	$K_{i\infty} = 2.07 \times 10^{-4} \rho_g (U_g - U_{ti}) Fr^\alpha Re_{ti}^{1.6} \left(\frac{\rho_p - \rho_g}{\rho_g} \right)^{0.61}$ <p>for Group A</p> $K_{i\infty} = 2.65 \times 10^{-6} \rho_g (U_g - U_{ti}) Fr^\alpha Re_{ti}^{1.6} \left(\frac{\rho_p - \rho_g}{\rho_g} \right)^{0.61}$ <p>$D_{col}^{-0.52} U_{mf}^{-0.65}$ for Group B</p> <p>$\alpha = Re_{ti}^{-0.4}$</p>	A, B; single species; column internals	2600, 2500, 840	37-252	0.2-1.1	Square bed (0.15x0.15)	
1987	Geldart and Wong [31]	$K_{i\infty} = 13.52 \rho_g U_g \exp\left(-30.3 \frac{U_{ti}}{U_g}\right)$	A, C; single species	364- 5000	5-95	(RH= 0-90 %)	0.08	2.6 (0.3)

1991	Sciazko et al. [62]	$K_{i\infty} = 1.6\rho_g \frac{U_g^2}{U_{ti}} \left(1 - \frac{U_{ti}}{U_g}\right)$	A, B; single species	1250	80-2000	0.51- 1.23	0.3	
1992	Baeyens et al. [27]	$K_{i\infty} = 5.4 \times 10^{-5} \rho_p \left(\frac{U_g}{0.2}\right)^{3.4} \left(1 - \frac{U_{ti}}{U_g}\right)^2$ for $d_{pi} < 10325\rho_p^{-0.725}$	A; multiple species	1320- 9442	10-78	0.21- 0.7	0.0762	2.72 (0.3- 0.35)
1994	Nakagawa et al. [22]	$K_{i\infty} = 2.5935 \times 10^{-3} \rho_p U_g R^{1.87} A^{0.55} H^{-0.64}$ $R = \sum x_i \frac{U_g - U_{ti}}{U_{ti}}$ for $U_{ti} < U_g$	A; single species	230- 1700	30-165	0.2-0.6	0.8, square bed (0.15x 0.15;	1-3 (0.3)

							0.08x0.08)	
1998	Tasirin and Geldart [7]	$K_{i\infty} = 23.7\rho_g U_g^{2.5} \exp\left(-\frac{5.4U_{ti}}{U_g}\right) \quad \text{for } Re_D < 3000$ $K_{i\infty} = 14.5\rho_g U_g^{2.5} \exp\left(-\frac{5.4U_{ti}}{U_g}\right) \quad \text{for } Re_D > 3000$	A	1180-1350	69-140	0.1-0.8	0.076, 0.152	2.4
1998	Ma and Kato [60]	$K_{i\infty} = 5.38\rho_g (U_g - U_{ti}) \left(\frac{d_{pi}(U_g - U_{ti})\rho_g}{\mu_g} \right)^{2.22}$ $\left(\frac{0.455\rho_p^{0.269}}{\rho_g d_{pi} g} \right)^{-0.15} \quad \text{for } d_{pi} < \frac{0.455\rho_p^{0.269}}{4.5g\rho_p}$	C, A, B; two species	2350, 2600	10-331	0.3-0.9 (RH<3 5 %)	0.071	1.02 (11-11.5)
1999	Choi et al. [46]	$K_{i\infty} = \frac{\mu_g Ar^{0.5} \exp\left(6.92 - 2.11F_g^{0.303} - \frac{13.1}{F_d^{0.902}}\right)}{d_{pi}}$ $F_g = g d_{pi} (\rho_p - \rho_g)$	A, B; multiple species	2400-6158	30-710	0.152-2.8 (T=293		1.97-9.1

		$F_d = \frac{C_d \rho_g U_g^2}{2}$ $C_d = \begin{cases} \frac{24}{Re_p} & \text{for } Re_p \leq 5.8 \\ \frac{10}{Re_p^{0.5}} & \text{for } 5.8 < Re_p \leq 540 \\ 0.43 & \text{for } Re_p > 540 \end{cases}$				-1173 K)		
1999	Tasirin and Geldart [68]	$K_{i\infty} = 2.66 \times 10^{-5} \rho_p \left(\frac{U_g}{0.2} \right)^3 \left(1 - \frac{U_{ti}}{U_g} \right)^{0.3} \text{ for } d_{pi} < d_{crit}$	A, C; single & multiple species		13-140 (6-16)	0.2-0.8 (RH< 35 %, P=4.4 atma)	0.076- 0.152	2.4
2000	Rodrigue z et al. [63]	$K_{i\infty} = \frac{\mu_g (U_g - U_{ti})^2 (1.5 \times 10^{-9} Re_{ti}^{0.6} + 2.5 \times 10^{-5} Re_{ti}^{1.2})}{g d_{pi}^2}$	C, B; multiple species	2481, 4799	37-210	0.19- 0.38	0.046	1

2001	Li and Kato [49]	$K_{i\infty} = 6.64 \times 10^6 C_{ps} \left(\frac{\rho_g U_g g}{\rho_p} \right)^{2.64} \text{ for } d_{pi} < d_{crit}$ $C_{ps} = \begin{cases} 1 & \text{for } d_{pi} \leq 60\mu m \\ \left(\frac{200 - d_{pi}}{150} \right)^\alpha + \left(\frac{d_{pi} - 60}{150} \right)^\alpha \left(\frac{d_{pi}}{d_{crit}} \right)^{1.4} & \text{for } 60\mu m \leq d_{pi} \leq 200\mu m \\ \left(\frac{d_{pi}}{d_{crit}} \right)^{1.4} & \text{for } d_{pi} \geq 200\mu m \end{cases}$ $\alpha = \left(\frac{d_{crit}}{d_{pi}} \right)^{0.3}, \quad d_{crit} = \frac{1.01 \times 10^5}{\rho_p g^{0.731}}$	C, A, B; multiple species	950- 3170	12-650	0.3-0.7 (RH=4 0-45 %)	0.103	2.55
2001	Choi et al. [35]	$K_{i\infty} = 0.314 C_d \rho_g U_g P^{0.946} \frac{F_d^{3.92}}{F_g^{2.04}}$ $F_g = g d_{pi} (\rho_p - \rho_g); \quad F_d = \frac{C_d \rho_g U_g^2}{2}; \quad P = \sum x (U_g - U_{ti})$	A, B; single species	2465- 2562	113-264	0.8-2.2	0.1	1.97 (0.15)

		$C_d = \begin{cases} \frac{24}{Re_p} & \text{for } Re_p \leq 5.8 \\ \frac{10}{Re_p^{0.5}} & \text{for } 5.8 < Re_p \leq 540 \\ 0.43 & \text{for } Re_p > 540 \end{cases}$						
2003	Stojkovski and Kostic [23]	$K_{i\infty} = \frac{K_1 \rho_g U_g \left(\frac{Fr_i^{1.125}}{Re_i} \right)^{K_2}}{\left(1 - \frac{U_{ti}}{U_g} \right)^{1.25}}$ $K_1 = 0.002518H^{-1.7170}; K_2 = 0.9997H^{0.2075}$	B; multiple species	2660	149-580 (0-100)	0.3-0.9 ($T=293$ -308 K)	0.2	3.26 (0.32)
2005	Chang et al. [69]	$K_{i\infty} = 0.0136 \rho_g U_{ti} \frac{Fr^{1.25}}{\left(\frac{T_{bed}}{T_{ambient}} \right)^{0.31}}$	C, B; single species	2850	40-97	0.4-1.5 ($T=920$ -1170 K)	0.2	2.5 (0.2)

2011	Han et al. [21]	$K_{i\infty}$ $= 0.968\rho_g U_{ti} \left[\frac{(U_g - U_{ti})^2}{gd_p} \right]^{0.878} \left(\frac{\rho_g d_p U_{ti}}{\mu} \right)^{0.432} \left(\frac{H}{D_c} \right)^{0.8}$	B	2420	81-545	0.9-1.8	0.19	3-5 (0.1)
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2.2. Results and Discussion

Assessment of the available entrainment correlations (Table 2.1) for continuous PSDs, monodisperse particles, and binary mixtures of Geldart Groups A and B particles will be carried out in this section. Because the ample number of correlations impedes readability of the plots, correlations that superimpose on one another are largely omitted from the figures and represented by a couple of similar plots.

2.2.1. Continuous PSDs particles

- **Geldart Group A particles**

This section presents the empirical predictions of total entrainment flux above TDH (K_∞) for continuous particle size distributions (PSDs), specifically for various widths of lognormal Group A particles. Furthermore, the elutriation of each size species (K_{iso}) in the PSD is examined.

The equation defining the mass-based frequency distribution (f_m) of a lognormal PSD is:

$$f_m(d_{pi}) = \frac{1}{d_{pi}\sigma_\mu\sqrt{2\pi}} \exp\left[-\frac{(\ln(d_{pi})-\mu)^2}{2\sigma_\mu^2}\right] \quad (1)$$

whereby d_{pi} represents characteristic particle diameter. The natural logarithm of this distribution is a Gaussian PSD with arithmetic mean μ and standard deviation σ_μ . In this work, the Sauter-mean diameter (d_{sm}) is kept constant, while the widths of the continuous PSDs, defined as the ratio of standard deviation of the PSD to sauter-mean diameter (σ/d_{sm}), are varied. The d_{sm} is used instead of other characteristic diameters since it is the most physically relevant; i.e., it preserves the ratio of volume to cross-sectional area, reflecting the downward gravitation pull and upward drag force in the suspension of a particle, respectively. As listed in Table 2.2, the values of d_{sm} and ρ_p

considered are 80 μm and 1450 kg/m^3 , respectively, which are common averages of fluid catalytic cracking (FCC) particles [70].

Table 2.2. Base case parameters for Geldart Group A lognormal PSDs.

Parameter	Symbol	Unit	Magnitude
Solid (FCC)	d_{sm}	μm	80
	ρ_p	kg/m^3	1450
	ϕ	-	0.58
	ε_{mf}	-	0.42
	U_t	m/s	0.13
Air	ρ_g	kg/m^3	1.2
	μ_g	kg/m/s	1.8×10^{-5}
Fluidizing unit	D_c	m	0.5
General	T	K	298
	g	kg/m/s^2	9.81

Accordingly, Fig. 2.1 presents the exact lognormal PSDs (equation (1)) as solid lines and corresponding size fractions discretized per standard sieve sizes, the latter of which is used in the calculation of K_∞ below. For the discrete points, f_m represents the mass fraction of each sieve cut (x_i) normalized with respect to the size difference between the given two sieves (i.e., Δd_{pi}). The widest PSD of $\sigma/d_{sm} = 0.70$ is such that particles still fall within Geldart Group A classification [24], which approximately is in the size range of 20-250 μm for the $\rho_p = 1450 \text{ kg/m}^3$ particles considered (Fig. 2.1).

Fig. 2.1. Lognormal PSDs of Geldart Group A particles investigated, with d_{sm} kept constant at $80 \mu\text{m}$ and σ/d_{sm} varied (see values in legend). The lines represent exact mass-based lognormal distributions as defined by equation (1), while the discrete points are discretized values used in calculating K_∞ .

The predicted K_∞ values for the narrowest ($\sigma/d_{sm} = 0.25$) and widest ($\sigma/d_{sm} = 0.70$) continuous PSDs of Group A particles at various ratios of superficial gas velocity to terminal velocity of the d_{sm} considered ($U_g/U_{t,80\mu\text{m}}$) are presented in Fig. 2.2, illustrating that the trends of K_∞ versus $U_g/U_{t,80\mu\text{m}}$ are largely similar for both PSDs. The range of $U_g/U_{t,80\mu\text{m}}$ is limited between 1.01 and 2.00, because the former represents the condition of incipient entrainment while the latter is the upper limit beyond which the solids begin to be entrained significantly. A noteworthy observation lies in that the discrepancies between the minimum and maximum predicted K_∞ values are orders of magnitude apart, with the largest being 12 orders of magnitude for $\sigma/d_{sm} = 0.70$ and $U_g/U_{t,80\mu\text{m}} = 2.00$. Expectedly, K_∞ generally increases as $U_g/U_{t,80\mu\text{m}}$ increases, specifically with K_∞ increasing up to 4 orders of magnitude as $U_g/U_{t,80\mu\text{m}}$ increases from 1.01 to 2.00 for both PSDs in Fig. 2.2. The increase in K_∞ with respect to $U_g/U_{t,80\mu\text{m}}$ of 4 orders of magnitude predicted by Kato et al. [59] is likely unphysical, despite the similarity between particle

system tested and the parameters specified here; however, the column cross-section used by Kato et al. [59] is squared instead of circular, which thereby puts forth a caveat in extrapolating predictions beyond the specific experimental setup. A second unphysical trend warrants mention too, namely, seven correlations [1, 28, 38, 57, 63, 65, 69] predict instead that K_∞ decreases as $U_g/U_{t,80\mu m}$ increases only for the widest PSD of $\sigma/d_{sm} = 0.70$ (Fig. 2.2b) but not for $\sigma/d_{sm} = 0.25$ (Fig. 2.2a), which indicates poorer predictive capability for wider PSDs. Even with the exclusion of the seven correlations [1, 28, 38, 57, 63, 65, 69] that have unphysical trends of K_∞ with respect to $U_g/U_{t,80\mu m}$ (namely, K_∞ decreases as $U_g/U_{t,80\mu m}$ increases), there still exists 12 orders of magnitude variations among the predicted K_∞ among the correlations for the PSD with $\sigma/d_{sm} = 0.70$. Furthermore, even if comparison is limited to correlations developed for Geldart Group A (see Table 2.1), the discrepancies among the correlations of the predicted K_∞ with respect to $U_g/U_{t,80\mu m}$ remain high at up to 11 orders of magnitude for the widest PSD considered ($\sigma/d_{sm} = 0.70$). This provides evidence again that correlations developed do not extrapolate well beyond the scope tested empirically.

Fig. 2.2. Total entrainment flux (K_{∞}) for lognormal Geldart Group A particles with

$\sigma/d_{sm} =$ (a) 0.25 and (b) 0.70 as a function of $U_g/U_{t,80\mu m}$.

To understand the effect of PSD width on K_{∞} values, it is necessary to evaluate the variations at both low and high $U_g/U_{t,80\mu m}$ values. Correspondingly, the total entrainment fluxes (K_{∞}) with respect to PSD width (σ/d_{sm}) are plotted in Fig. 2.3 for two values of $U_g/U_{t,80\mu m}$, namely 1.01 and 2.00. A few observations are worth noting in Fig. 2.3. First, predicted K_{∞} values are orders of magnitude apart among the various correlations. In particular, differences between the minimum and maximum K_{∞} values predicted are in the range of 7 and 13 orders of magnitude at the lower and higher $U_g/U_{t,80\mu m}$, respectively. Second, the bulk of correlations have better agreement at higher $U_g/U_{t,80\mu m}$ (Fig. 2.3b). Third, a general lack of predictive ability of changes in K_{∞} with σ/d_{sm} seems apparent, based on previous experimental results (for Group B particles) which indicated that K_{∞} increases by 150 % for a $\sigma/d_{sm} = 0.70$ PSD relative to that for

monodisperse (akin to $\sigma/d_{sm} = 0.1$) at $U_g/U_{t,dsm} = 1.06$ [15]. While only two correlations [19, 49] predict a slight negative correlation between K_∞ and σ/d_{sm} at the lower $U_g/U_{t,80\mu m}$ (Fig. 2.3a), seven [19, 45, 49, 58, 60, 66, 68] so predict at the higher $U_g/U_{t,80\mu m}$ (Fig. 2.3b). Furthermore on variations of K_∞ between the narrowest ($\sigma/d_{sm} = 0.1$) and widest ($\sigma/d_{sm} = 0.7$) PSDs, for the lower $U_g/U_{t,80\mu m}$ (Fig. 2.3a), only six correlations give a difference of K_∞ of within 300 % [13, 36, 37, 45, 49, 68], while thirteen correlations [1, 27, 28, 31, 35, 38, 46, 57, 59, 60, 63, 65, 69] predict a four or more orders of magnitude change, with two predicting more than 10 orders of magnitude [31, 59]. As for the higher $U_g/U_{t,80\mu m}$ (Fig. 2.3b), twenty one [1, 13, 14, 27, 28, 35-38, 45, 49, 57, 58, 60, 62, 63, 65-69] correlations give a difference of K_∞ of up to 300 %, with Kato et al. [59] further predict a colossal 8 orders of magnitude of change. Fourth, although it may be expected that correlations based on data from Geldart Group A particles may give more similar predictions of K_∞ values, it is not the case since correlations for Geldart Group B or a crossover among the Geldart Groups overlap in the same range.

Fig. 2.3. Total entrainment flux (K_∞) for Geldart Group A particles as a function of

σ/d_{sm} at $U_g/U_{t,80\mu m}$ of (a) 1.01 and (b) 2.00.

To further understand elutriation characteristics, the elutriated PSDs derived via the correlations in Table 2.1 are displayed in Fig. 2.4, which are plots of f_m versus d_p at two different $U_g/U_{t,80\mu m}$ values. Interestingly, for a PSD with $\sigma/d_{sm} = 0.70$ at $U_g/U_{t,80\mu m} = 1.01$ (Fig. 2.4a), the PSDs predicted by the correlations can be categorized rather evenly into three groups, namely, (i) dominance of smallest particles (five correlations [31, 35, 59, 67, 69]), (ii) continuous PSDs skewed leftwards with respect to the initial bed PSD (eleven correlations [7, 13, 14, 36, 37, 45, 58, 60, 62, 66, 68]), and (iii) continuous PSDs skewed rightward with respect to the initial bed PSD (one correlation [49]) and dominance of larger particles (eight correlations [1, 27, 28, 38, 57, 63, 65, 69]). Thus, in addition to the enormous quantitative difference in total elutriation flux illustrated in Fig. 2.3, Fig. 2.4 demonstrates a striking qualitative difference in the correlation predictions for the shape of elutriated PSD. Regarding (i), the inflated representation of the smallest particles in the elutriated PSD is physically reasonable, since the associated U_{ti} (terminal velocity of particles with size d_{pi}) is much smaller than the operating U_g , and hence smallest particles have the highest propensity to be entrained, although it has also been reported that fine particles were not always the most likely to be entrained from a bed of mixed particle sizes [56, 71]. Also, in this category, the inherent assumption is that particles with $U_{ti} > U_g$ cannot be entrained, although experimental data for both Geldart Group A and B particles indicates the contrary, namely that collisional momentum transfer between smaller and larger particles enables entrainment of the latter [14, 15]. The leftward shift of the elutriated PSD in category (ii) also is physically sound, stemming from the same reasoning as (i). In this category, some

correlations predict that only particles whose $U_{ti} < U_g$ are elutriated, whereas three display more prominently elutriation of particles whose $U_{ti} > U_g$ [13, 36, 45]. An exception in this category is that of George and Grace [37], which predicts that the elutriated PSD is the same as that in the bed since the correlation is only dependent on U_g and regardless of any particle property. Category (iii) contains unphysical trends, specifically that larger particles are featured prominently in the elutriated PSD. Upon closer inspection of the correlations (Table 2.1), it is observed that a common term is either $(U_g - U_{ti})^2$ or $(1 - \frac{U_g}{U_t})^2$, which thus instead of accounting for the negative $(U_g - U_{ti})$ or $(1 - \frac{U_g}{U_t})$ associated with the larger particles, the amount of larger particles elutriated is instead magnified. The only correlation in this category without either of these two terms is Li and Kato [49]; this correlation was targeted at cohesive Group C particles, thus the erroneous higher elutriation of the larger particles may be rooted in the *ad hoc* inclusion of empirical constants to inflate the K_∞ value to account for inter-particle cohesive effects.

Fig. 2.4. Elutriated PSDs for Geldart Group A particles with initial bed PSD of $\sigma/d_{sm}=0.70$ at $U_g/U_{t,80\mu m}$ of (a) 1.01 and (b) 2.00.

The similar three categories of correlations persist for the PSD with $\sigma/d_{sm}=0.70$ at the higher $U_g/U_{t,80\mu m}$ of 2.00 (Fig. 2.4b). Compared to that for Fig. 2.4a, category (i) contains fewer correlations [35, 59, 67] which predict the prevalence of the smallest particles in the elutriated. Category (ii) is similarly a leftward shift of the elutriated PSD relative to the bed PSD. With regards to category (iii) noted above (Fig. 2.4a), the term $(U_g - U_{ti})^2$ or $(1 - \frac{U_g}{U_t})^2$ remains a key factor, though it becomes less negative as U_g increases, and hence the anomalous inflation of larger particles is somewhat attenuated vis-à-vis the lower $U_g/U_{t,80\mu m}$ condition (Fig. 2.4a). Interestingly, out of the eight correlations that predict a rightward shift of the elutriated PSD with respect to the initial bed PSD, half of them [1, 27, 28, 38] were developed for Group A, which thereby implies that the restriction of correlations to the Geldart Group they were developed for does not seem to enhance the prediction of the elutriation phenomenon.

For the narrower lognormal PSD of $\sigma/d_{sm} = 0.25$ portrayed in Fig. 2.5, three interesting observations warrants highlighting. First, the anomalous elevated elutriation of the largest particles seen in Fig. 2.4a is absent at both $U_g/U_{t,80\mu m}$ values, although the rightward shift of the elutriated PSD relative to the bed PSD is still predicted by some correlations at both $U_g/U_{t,80\mu m}$ values. Second, at the lower $U_g/U_{t,80\mu m}$ (Fig. 2.5a), the

elutriated PSDs can be classified into three categories: (i) elutriation of the smallest particles [59] (ii) leftward shift with respect to initial bed PSD, and (iii) bimodal elutriated PSD. More specifically for (iii), eight correlations predict elutriated PSDs with two peaks at $54\ \mu\text{m}$ and $115.5\ \mu\text{m}$ (note $U_{t,54\mu\text{m}} < U_g$ and $U_{t,115.5\mu\text{m}} > U_g$) particles[1, 27, 28, 38, 57, 63, 65, 69], the lack of physical soundness of which can similarly be traced to the term $(U_g - U_t)^2$ or $(1 - \frac{U_g}{U_t})^2$ as elucidated above. Third, for the $U_g/U_{t,80\mu\text{m}} = 2$ condition (Fig. 2.5b), save for Kato et al. [59], the shapes of the elutriated PSDs are largely similar to one another and approximately preserved with respect to the starting bed PSD. Combined, Figs. 2.4 – 2.5, reflecting elutriated PSDs from narrow and wide bed PSDs, respectively, highlight two important observations: first, even though d_{sm} is kept constant, the impact of varying widths of PSDs cannot be ignored, especially if the nature of the elutriated PSDs is significant enough to affect a process; and second, the pitfalls involved in the application of available correlations to systems beyond the specific system used for the development of the correlations.

Fig. 2.5. Elutriated PSDs for Geldart Group A particles with initial bed PSD of $\sigma/d_{sm} = 0.25$ at $U_g/U_{t,80\mu m}$ of 1.01 and (b) 2.00.

• Geldart Group B particles

Similar to the previous section on Group A particles, the elutriation correlations will now be compared for Group B particles with lognormal PSDs. Listed in Table 2.3 are the values of d_{sm} and ρ_p considered for Geldart Group B particles, namely 325 μm and 2500 kg/m^3 , respectively. Analogous to Fig. 2.1 for Geldart Group A, Fig. 2.6 displays the exact lognormal PSDs (Equation 1) for Geldart Group B particles. The widest PSD ($\sigma/d_{sm} = 0.70$) is such that particles still largely fall within Geldart Group B classification [24], which approximately is in the size range of 80–700 μm for the $\rho_p = 2500 kg/m^3$ particles considered.

Table 2.3. Base case parameters for Geldart Group B lognormal PSDs.

Parameter	Symbol	Unit	Magnitude
Solid (glass)	d_{sm}	μm	325
	ρ_p	kg/m^3	2500
	Φ	-	0.95
	ε_{mf}	-	0.42
	U_t	m/s	2.58

Air	ρ_g	kg/m ³	1.2
	μ_g	kg/m/s	1.8×10^{-5}
Fluidizing unit	D_c	M	0.5
General	T	K	298
	G	kg/m/s ²	9.81

Fig. 2.6. Lognormal PSDs of Geldart Group B particles investigated, with d_{sm} kept constant at 325 μm and σ/d_{sm} varied. The lines represent exact mass-based lognormal distributions as defined by equation (1), while the discrete points are discretized values used in calculating K_∞ .

Analogous to Fig. 2.2, Fig. 2.7 depicts the predicted K_∞ values for the narrowest ($\sigma/d_{sm} = 0.25$) and widest ($\sigma/d_{sm} = 0.70$) lognormal PSDs of Group B particles at various $U_g/U_{t,325\mu\text{m}}$ values. Notably, the outlying lowermost trend in Fig. 2.7a is developed for Geldart Group A and C particles by Geldart and Wong [31], thus the application here to Group B particles may seem unjust; however, three other correlations not developed for Group B [7, 13, 27] are indistinguishable from those developed for Group B, hence indicating that whether a particular correlation was developed for a specific Geldart

Group seems inconsequential to giving better predictions even when appropriately applied to the particular subset. Discrepancies between minimum and maximum predicted K_∞ values are in the range of 6 and 11 orders of magnitude, which is lower than that for Group A (Fig. 2.2). As expected, K_∞ generally increases with $U_g/U_{t,80\mu m}$, with the largest increase of K_∞ in the $U_g/U_{t,325\mu m}$ range considered of 3 orders of magnitude for the narrower PSD ($\sigma/d_{sm} = 0.25$) [31, 60], which is also lower than that for Group A. Moreover, unlike for Group A whereby K_∞ decreases as $U_g/U_{t,80\mu m}$ increases for the widest PSD, K_∞ consistently increases with $U_g/U_{t,325\mu m}$ for Group B. When comparisons are limited only to the correlations that were developed for Group B (Table 2.1), discrepancies among predicted K_∞ are also smaller at between 4 to 6 orders of magnitude (for the PSDs with $\sigma/d_{sm} = 0.25$ and $\sigma/d_{sm} = 0.70$) compared to that for polydisperse Group A. Thereby, relative to Group A, the predictions for Group B are somewhat less unphysical (i.e., discrepancies among Geldart B correlation are 5 orders of magnitude less than that of Geldart A correlations).

Fig. 2.7. Total entrainment flux (K_∞) for lognormal Geldart Group B particles with

$\sigma/d_{sm} =$ (a) 0.25 and (b) 0.70 as a function of $U_g/U_{t,325\mu m}$.

Analogous to Fig. 2.3 for Geldart Group A, the total entrainment fluxes (K_∞) with respect to PSD width (σ/d_{sm}) for Geldart Group B are plotted in Fig. 2.8 for the same values of $U_g/U_{t,325\mu m}$ of 1.01 and 2. Unsurprisingly, discrepancies among predicted K_∞ are huge, namely between 5 and 16 orders of magnitude apart among the various correlations. Similar to that for Group A (Fig. 2.3), disagreements with experimental results [14, 15] in terms of negative correlation between K_∞ and σ/d_{sm} is prevalent at the higher $U_g/U_{t,325\mu m}$.

Fig. 2.8. Total entrainment flux (K_∞) for Geldart Group B particles as a function of

σ/d_{sm} at $U_g/U_{t,325\mu m}$ of (a) 1.01 and (b) 2.00.

Three differences between Geldart Group B (Fig. 2.8) and A (Fig. 2.3) are worth noting. First, in contrast to Group A whereby the largest order of magnitude difference among predictions is 13 for the *widest* PSD ($\sigma/d_{sm} = 0.70$) at the *higher* $U_g/U_{t,dsm}$ (Fig. 2.3), the largest order of magnitude difference is larger at 16 and occurs for the *narrowest* PSD ($\sigma/d_{sm} = 0.10$) at the *lower* $U_g/U_{t,dsm}$ (Fig. 2.8a). Second, for the same $U_g/U_{t,dsm}$, Group B particles generally have higher K_∞ values than Group A. Third, fewer correlations exhibit drastic variations of K_∞ with σ/d_{sm} , with only four correlations [27, 31, 60, 69] giving variations of four or more orders of magnitude at the lower $U_g/U_{t,325\mu m}$, and only two correlations [31, 35] predicting a greater than 300 % change at the higher $U_g/U_{t,325\mu m}$. In particular, the K_∞ values predicted by Kato et al.[59] and Choi et al.[46], both developed for Geldart Groups A and B, are relatively flat as σ/d_{sm} increases for Group B, compared to the steep increase of K_∞ with σ/d_{sm} for Group A. Moreover, at the higher $U_g/U_{t,325\mu m}$ (Fig. 2.8b), all K_∞ values are relatively invariant with respect to σ/d_{sm} except for the Geldart and Wong [31] correlation, presumably because this correlation was developed for fine particles and thus the increased inter-particle cohesive effects considered are not applicable for the Geldart Group B considered here. Regarding commonality between Group A and Group B, more correlations give large variations of K_∞ with respect to σ/d_{sm} at the lower $U_g/U_{t,dsm}$ value compared to the higher one.

Regarding elutriated PSDs for the widest lognormal PSD considered ($\sigma/d_{sm} = 0.70$), the shapes of the PSDs for Group B (Fig. 2.9) generally resemble those for Group A (Fig. 2.4). For the lower $U_g/U_{t,325\mu m}$, the shapes of the PSDs also fall into three categories, but a few noteworthy differences between Group A (Fig. 2.4a) and B (Fig. 2.9a) at the lower $U_g/U_{t,dsm}$ are observed. For the first category whereby the smallest particles dominate, of the five correlations for Group A, only four exhibit this trend for Group B [31, 35, 59, 67], the exception being Choi et al. [46] (developed for both

Geldart Groups A and B) moving into the second category. For the third category described above for Group A which exhibit the dominant presence of the largest particles (Fig. 2.4a), a differentiation for Group B (Fig. 2.9a) is apparent in that an additional distinct peak exists for the intermediate small particles ($d_{pi} = 165 \mu\text{m}$). In particular, all of the eight correlations referenced above for Group A displaying the prominent representation of the largest particles ($215 \mu\text{m}$) predict a similar trend here, with the exception of the distinctly higher representation of the intermediate small particles ($165 \mu\text{m}$) for Group B. Surprisingly, although it was expected that correlations developed for Group B particles would have better predictive capability here, seven [1, 28, 38, 57, 63, 65, 69] out of the eight correlations that predict the unphysical rightward shift of elutriated PSDs were developed for Group B. Regardless of whether the correlations are applied to Group A or B systems, the non-physical preferential elutriation of the largest particles is predicted at the lower $U_g/U_{t,dsm}$.

Fig. 2.9. Elutriated PSDs for Geldart Group B particles with initial bed PSD of $\sigma/d_{sm} = 0.70$ at $U_g/U_{t,325\mu m}$ of (a) 1.01 and (b) 2.00.

Fig. 2.9b shows the elutriated PSDs predicted at the higher $U_g/U_{t,325\mu m}$ of 2. Comparing with the analogous chart for Group A (Fig. 2.4b), one similarity and one notable difference are worth highlighting. The similarity lies in that the three correlations that predict the dominant elutriation of the smallest particles (19 μm for Group A and 75 μm for Group B) are the same ones for both groups. As for the difference between the groups, although numerous correlations predict the unphysical preferential elutriation of the larger particles in Group A (Fig. 2.4b), no correlation predict the dominance of largest particles in the elutriation of Group B PSD. Specifically, only Li and Kato [49] predict a slightly rightward shift of elutriated PSD, while all other correlations predict a leftwards shift of the elutriated PSD relative to the initial bed PSD in the case of Group B (Fig. 2.9b).

As for elutriated PSDs for lognormal PSD with $\sigma/d_{sm} = 0.25$, Group A (Fig. 2.5) and Group B (Fig. 2.10) exhibit largely similar trends, except that the dominant elutriation of the smallest particles [59] exists only for Group A (Fig. 2.5) but not Group B (Fig. 2.10) at both $U_g/U_{t,dsm}$ values. One similarity is that, at the lower $U_g/U_{t,dsm}$, the same eight correlations [1, 27, 28, 38, 57, 63, 65, 69] predict elutriated PSDs with two peaks for both Groups A (Fig. 2.5a) and B (Fig. 2.10a). Again, seven [1, 28, 38, 57, 63, 65, 69] out of the eight correlations predicting the unphysical trends were actually developed for Group B particles, which highlights once more the lack of predictive capability of empirical correlations even if applicability is restricted to the Geldart Group the correlations were developed for. Specifically, the two peaks for in the case for Group B are at 231 μm and 450 μm (note $U_{t,231\mu m} < U_g$ and $U_{t,450\mu m} > U_g$). The other

similarities between Group A (Fig. 2.5b) and Group B (Fig. 2.10b) at the higher $U_g/U_{t,dsm}$ are that the bimodality of the elutriated PSDs is absent and shapes of elutriated PSDs are generally preserved with respect to the initial bed PSD. It is worth mentioning that the objective of Fig. 2.9 and 2.10 is to invoke awareness among fluidization community about the huge discrepancies of entrainment flux predicted by existing correlations (even within their validity range), and the need of a mechanistic study on fluidization phenomenon to avoid the use of empirical constants to fir particular experimental data set and design correlation. Furthermore, such entrainment correlations do not account for the impact of particle polydispersity, which is shown to be influential on affecting entrainment flux.

Fig. 2.10. Elutriated PSDs for Geldart Group B particles with initial bed PSD of $\sigma/d_{sm} = 0.25$ at $U_g/U_{t,325\mu\text{m}}$ of (a) 1.01 and (b) 2.

2.2.2. Monodisperse particles

- **Geldart Group A particles**

Equipped with an understanding of the elutriation trends for lognormal PSDs from the previous sections, it is expedient to further examine the predictions of K_{∞} values for the constituent monodisperse particles. Accordingly, this section and the next explores the entrainment trends for monodisperse Group A and B particles, respectively. For monodisperse Group A, Table 2.4 lists the base case parameters considered, specifically for d_{pi} in the range of 25 to 200 μm .

Table 2.4. Base case parameters for monodisperse Geldart Group A particles.

Parameter	Symbol	Unit	Magnitude
Solid (FCC)	d_{pi}	μm	25 – 200
	ρ_p	kg/m^3	1450
	Φ	-	0.58
	ε_{mf}	-	0.42
	U_t	m/s	0.015 - 0.48
Air	ρ_g	kg/m^3	1.2
	μ_g	kg/m/s	1.8×10^{-5}
Fluidizing unit	D_c	m	0.5
General	T	K	298
	G	kg/m/s^2	9.81

The entrainment trends for monodisperse 80 μm FCC particles are displayed in Fig. 2.11 at various $U_g/U_{t,80\mu\text{m}}$ (Fig. 2.11a) and $U_g - U_{t,80\mu\text{m}}$ (Fig. 2.11b) values, because many correlations are functions of either the former or latter. Two notes on the

distinction between U_g/U_{ti} and $U_g - U_{ti}$ are worth highlighting: (i) It is worth noting that the former is dimensionless, whereas the latter has unit of m/s, and (ii) at fixed U_g/U_{ti} , $U_g - U_{ti}$ increases as d_{pi} (and hence U_{ti}) increases. Four observations merit attention. First, along the same vein as previously noted, predicted K_∞ values are orders of magnitude apart among the various correlations. Notably, in the ranges of $U_g/U_{t,80\mu m}$ (Fig. 2.11a) and $U_g - U_{t,80\mu m}$ (Fig. 2.11b) considered, the order of magnitude difference between the minimum and maximum values of K_∞ decreases monotonically from 13 to 6. Also, the largest variations of predicted K_∞ occurs at the lowest $U_g/U_{t,80\mu m}$ (Fig. 2.11a) or $U_g - U_{t,80\mu m}$ (Fig. 2.11b). Second, in both subplots in Fig. 2.11, K_∞ increases monotonically as either $U_g/U_{t,80\mu m}$ or $U_g - U_{t,80\mu m}$ increases, with the exception being Matsen [67], which decreases slightly at the lower end of $U_g/U_{t,80\mu m}$ and $U_g - U_{t,80\mu m}$ before increasing at higher $U_g/U_{t,80\mu m}$ and $U_g - U_{t,80\mu m}$. The reason for this slight aberration by Matsen [67] is the hypothesized demarcation of flow phenomenon based on the slip velocity – voidage relationship between choking ($U_g/U_{t,80\mu m} < 1.29$) and non-choking ($U_g/U_{t,80\mu m} > 1.29$) scenarios. Third, comparing Fig. 2.2 for lognormal Group A PSDs and Fig. 2.11 for monodisperse Group A particles, the trends are largely similar. Geldart and Wong [31], Choi et al. [46] and Rodriguez et al. [63] consistently predict the lowest K_∞ values in both monodisperse and polydisperse cases; however, Kato et al. [59] gives the outlying highest K_∞ values for the widest PSD (Fig. 2.2b) but not for the narrower PSD (Fig. 2.2a) nor monodisperse system (Fig. 2.11). In particular, for the Kato et al. correlation [59], it was developed for continuous PSDs, which makes it surprising that the predicted K_∞ values are outliers for the polydisperse (Fig. 2.2) but not the monodisperse (Fig. 2.11), although being outlying by no means suggest whether the prediction is more or less accurate for the system considered here. Nonetheless, a plausible implication is that the interactions among various size species are unique to

specific systems, hence extrapolation of such empirical correlations beyond tested parameters may lead to erroneous predictions. Four, the trends for $U_g/U_{t,80\mu m}$ (Fig. 2.11a) and $U_g - U_{t,80\mu m}$ (Fig. 2.11b) are largely similar, although the range of values of $U_g/U_{t,80\mu m}$ and $U_g - U_{t,80\mu m}$ used are necessarily different.

Fig. 2.11. Total entrainment flux (K_∞) for 80 μm FCC particles (Group A) as a function of (a) $U_g/U_{t,80\mu m}$, and (b) $U_g - U_{t,80\mu m}$.

Accordingly, the physical basis underlying the lowest and highest predicted K_∞ values can be explored to gain more understanding towards a more physically-based model for entrainment. Regarding the lower end of K_∞ values in Fig. 2.11, Geldart and Wong [31], Choi et al. [46], and Rodriguez et al. [63] give the outlying lowest K_∞ values across the range of $U_g/U_{t,80\mu m}$ and $U_g - U_{t,80\mu m}$ considered. Upon closer inspection of the references, each presents a different physical reasoning, all of which are sound. For Choi et al. [46], the term *dispersed non-cluster flux* was construed, and the correlation was

applicable to such category of Group B particles, hence the explicit neglecting of cluster flux leads to lower K_∞ , with the discrepancy presumably made up of cluster flux. Evident per Cocco et al. [72], clustering is significant for both the FCC (Group A) and polyethylene (Group B) particles examined through high-speed video imaging. As for Geldart and Wong [31] and Rodriguez et al. [63], elutriation of binary mixtures of Group A and Group C particles was studied, and it was hypothesized that the decrease in K_∞ is due to inter-particle interactions, which is very prominent in Group C; thereby, extrapolation to Group A only in this case results in low prediction in K_∞ presumably due to over-consideration of inter-particle cohesion forces, which hints that correlations so developed for Group C may be too system-specific to be amenable for other groups. However, a few other correlations that investigated Group C particles too [36, 49, 60, 69] present different results. For example, Lin et al. [36] instead gives higher K_∞ at the higher $U_g - U_{t,80\mu m}$ (Fig. 2.11b). Taken together, although the particles studied in Geldart and Wong [31], Choi et al. [46], and Rodriguez et al. [63] belong to dissimilar Geldart groups, the common thread for the lower K_∞ values predicted lies in the accounting of inter-particle interactions, either leading to cohesive agglomerates (commonly observed for Group C and smaller Group A particles) or dynamic clusters (due to hydrodynamic instabilities), which thereby implies such effects needs further understanding towards better predictions of entrainment.

With regards to the highest K_∞ values in Fig. 2.11, at the lowest $U_g/U_{t,80\mu m}$ and $U_g - U_{t,80\mu m}$, Merrick and Highley [45], Li and Kato [49], and Matsen [67] give the three highest K_∞ values. Closer inspection of the three references found: (i) Merrick and Highley [45] uniquely contains the term $(U_g - U_{mfi})$ in the correlation; the only other correlation inclusive of U_{mfi} is Kato et al. [59] targeted at Group B particles, but the K_∞ so predicted is not outlying, (ii) Merrick and Highley [45] and Li and Kato [49]

attempted to incorporate the effects of particle attrition, which may not be readily extrapolated to systems beyond that tested, (iii) The Li and Kato [49] correlation was developed for elutriation of the fines component (Group A and C) from binary mixtures (Group A, B, and C), which thereby also alludes to the possibility of mis-accounting of inter-particle interactions when extrapolated to a monodisperse system in this case, and (iv) Matsen [67] hypothesized that slip velocity is elevated in denser systems, thus inflated K_{∞} to correct for lower-velocity systems ($U_g/U_t < 1.29$). Seemingly, (ii) - (iv) infer that, without a universal way of incorporating inter-particle effects (e.g., attrition and cohesion), correlations may only be applicable for the specific systems studied. On the other hand, at the highest $U_g/U_{t,80\mu m}$ and $U_g - U_{t,80\mu m}$, Colakyan et al. [65], Merrick and Highley [45], and Li and Kato [49] give the three highest K_{∞} values. Notably, Li and Kato [49] was developed for Group A and C particles, hence presumably the inter-particle cohesive effects were over-accounted for when applied to the 80 μm FCC particles (Group A) considered here. On the other hand, Merrick and Highley [45] was developed to predict the elutriation of a wide PSD of coal in the presence of fine particles, while Colakyan et al. [65] was developed to predict elutriation of large particles (Group B particles). Apparently, at higher U_g values, with reference to Merrick and Highley [45] and Colakyan et al. [65], the accounting of the effects of polydispersity led to inflation of K_{∞} values when applied to a monodisperse system in this case. Notably, most correlations (Table 2.1) were developed for systems containing multiple species; hence, it is plausible that the lack of understanding of the interactions among different species may lead to the lack of capability for application to other systems. Therefore, regarding the outlying correlations in Fig. 2.11, the underlying physical basis lies in either inter-particle interactions (resulting in either cohesive agglomeration or hydrodynamic clustering) or polydispersity (leading to phenomenon like collisional momentum

transfer among species), which thereby implies a necessity for enhanced understanding of these phenomena.

Furthermore, analogous to Fig. 2.3 and Fig. 2.8 for the lognormal PSDs, Fig. 2.12 explores the variations of K_∞ values predicted by various correlations (Table 2.1) as a function of d_{pi} at $U_g/U_{ti} = 1.01$ (Fig. 2.12a) and 2 (Fig. 2.12b). Regarding discrepancies among correlations, at the lower U_g/U_{ti} (Fig. 2.12a), orders of magnitude difference in predicted K_∞ values is up to 20 at the lowermost d_{pi} in Group A, and decreases to 14 at the highest limit of d_{pi} ; on the other hand, at the higher U_g/U_{ti} (Fig. 2.12b), the lowest order of magnitude difference is 5 at an intermediate particle size of 80 μm , and between 7 and 9 for other d_{pi} values. The vast variations of K_∞ values indicate the lack of applicability of existing correlations beyond the systems studied, hence the only reasonable option is a fundamental description.

Generally, at fixed U_g/U_{ti} , K_∞ increases as d_{pi} increases (Fig. 2.12). Two exceptions are noted: (i) Three correlations [38, 65, 67] stipulate that K_∞ is invariant with d_{pi} , because U_g/U_{ti} is the only parameter taken into account, and (ii) Kato et al. [59], which proposed a correlation each for Group A and B, stipulates that K_∞ decreases then increases with d_{pi} only at the higher U_g/U_{ti} . By excluding the four correlations [38, 59, 65, 67] that give unphysical trends of K_∞ with respect to d_{pi} , there still exists between 5 and 19 orders of magnitude variations between the minimum and maximum predicted K_∞ values. When comparisons are limited only to correlations developed for Group A (Table 2.1), the discrepancies among the minimum and maximum K_∞ values predicted are still enormous at between 5 and 19 orders of magnitude. Notably, the maximum order of magnitude increase in K_∞ as d_{pi} increases in the range considered are immense at 18 [46] and 9 [46] at the lower and higher U_g/U_{ti} , respectively, which if true will inevitably lead to process upsets. In particular, at the lower U_g/U_{ti} , Choi et al. [46]

predict the greatest increase in K_∞ as d_{pi} increases from 25 to 200 μm , because of the explicit lack of consideration for inter-particle cohesion, the attentive correction of which will lead to over-prediction of K_∞ at higher d_{pi} values wherein inter-particle cohesion is lower. Again, the *ad hoc* inclusion or exclusion of some parameters over others in these empirical correlations proves inadequate towards accurate predictions of K_∞ .

Fig. 2.12. Total entrainment flux (K_∞) for FCC particles (Group A) as a function of d_{pi} at $U_g/U_{ti} =$ (a) 1.01 and (b) 2.

• Geldart Group B particles

This section addresses the predictions of K_∞ values for the constituent monodisperse particles belonging to Group B. Listed in Table 2.5 is the base case parameters considered, specifically for d_{pi} in the range of 80 to 700 μm .

Table 2.5. Base case parameters for monodisperse Geldart Group B particles.

Parameter	Symbol	Unit	Magnitude
Solid (glass)	d_{pi}	μm	80 – 700
	ρ_p	kg/m^3	2500
	Φ	-	0.95
	ε_{mf}	-	0.42
	U_t	m/s	0.39 - 4.8
Air	ρ_g	kg/m^3	1.2
	μ_g	kg/m/s	1.8×10^{-5}
Fluidizing unit	D_c	m	0.5
General	T	K	298
	G	kg/m/s^2	9.81

Analogous to Fig. 2.11, which illustrates entrainment trends for Group A particles across ranges of the commonly seen terms of U_g/U_t and $U_g - U_t$ in the correlations (Table 2.1), Fig. 2.13 presents the entrainment trends for monodisperse glass beads with $d_{pi} = 325 \mu\text{m}$ (Group B) in the same range of U_g/U_t (Fig. 2.13a) and $U_g - U_t$ (Fig. 2.13b) values. The largest discrepancies among the predictions is similar at a stunning 16 orders of magnitude both at the lowest end of $U_g/U_{t,325\mu\text{m}}$ and $U_g - U_{t,325\mu\text{m}}$, respectively. In particular, Geldart and Wong [31] and Rodrigues et al. [63], which were developed for cohesive Geldart Group A and C, generally gives the outlying lowest K_∞ predictions in the ranges of $U_g/U_{t,325\mu\text{m}}$ and $U_g - U_{t,325\mu\text{m}}$ considered. Furthermore, K_∞ increases monotonically as either $U_g/U_{t,325\mu\text{m}}$ or $U_g - U_{t,325\mu\text{m}}$ increases, with the exception being Matsen [67], which displays a non-monotonic behavior due to the

consideration of the choking transition at $U_g/U_{t,325\mu m} = 1.29$. The largest change in order of magnitude in K_∞ values in the ranges of $U_g/U_{t,325\mu m}$ and $U_g - U_{t,325\mu m}$ considered are 6 [60] and 8 [60]. Notably, Ma and Kato [60] were developed for Geldart Group A and C, hence again implies the lack of comprehensive understanding of inter-particle interactive effects impedes extrapolation of existing correlations to systems beyond the scope tested.

Fig. 2.13. Total entrainment flux (K_∞) for 325 μm glass beads (Group B) as a function of (a) $U_g/U_{t,325\mu m}$ and (b) $U_g - U_{t,325\mu m}$.

Fig. 2.14 in turn displays the entrainment trends for monodisperse glass beads at two fixed U_g/U_{ti} values as functions of d_{pi} values within the Group B classification.

Fig. 2.14. Total entrainment flux (K_{∞}) for glass beads (Group B) as a function of d_{pi} at

$U_g/U_{ti} = (a) 1.01$ and $(b) 2.00$.

The largest order of magnitude discrepancies between the minimum and maximum K_{∞} values, found at the highest d_{pi} values, are 17 and 11 in Figs. 2.14a and b, respectively. At $U_g/U_{ti} = 1.01$ (Fig. 2.14a) and $U_g/U_{ti} = 2.00$ (Fig. 2.14b), the lowest K_{∞} values are once more predicted by Geldart and Wong [31], while the highest K_{∞} values are predicted by Li and Kato [49]; despite the huge discrepancies, the common thread for these two correlations is that Geldart Groups A and C particles were investigated. Moreover, generally, K_{∞} increases as d_{pi} increases at fixed U_g/U_{ti} , with K_{∞} increasing up to 5 and 8 orders of magnitude in the range of d_{pi} considered at the lower and higher U_g/U_{ti} , respectively. The three exceptions predicting that K_{∞} is invariant with d_{pi} are such that K_{∞} is only dependent on ρ_p and/or the U_g/U_{ti} ratio [38, 65, 67]. By excluding these three correlations [38, 65, 67] that give unphysical trends of K_{∞} with respect to d_{pi} , the

discrepancies between the minimum and maximum K_{∞} values predicted are lowered but still high at 7 and 17 orders of magnitude at $U_g/U_{t,325\mu m}=1.01$ (Fig. 2.14a) and $U_g/U_{t,325\mu m}=2.00$ (Fig. 2.14b), respectively. When correlations considered are limited only to those developed for Group B (Table 2.1), the discrepancies between the minimum and maximum K_{∞} values predicted remain high at 4 to 9 orders of magnitude.

2.2.3. Binary mixtures

- **Geldart Group A particles**

Other than the polydisperse systems of continuous PSDs discussed earlier, binary systems are also common. Hence, these couple of sections addresses entrainment predictions (Table 2.1) for binary systems, specifically consisting of two particles differing only in size (d_{pi}) or only in particle density (ρ_p). In order to allow for fairer comparison between the two types of binary systems, which have been shown to behave differently [15, 73-77], the size and density binary systems for each Group are chosen such that the ratios of U_{ti} values between the two species are similar.

For the size binary system of Group A particles, Table 2.6 lists the base case parameters. Specifically, the system consists of two sizes (d_{pi}), namely 40 μm and 75 μm , of FCC particles. The ratio of U_{ti} values between the finer and coarser FCC particles is approximately 2.97. Accordingly, Fig. 2.15 displays the entrainment trends from available correlations (Table 2.1) as functions of mass fraction of the finer 40 μm FCC at two different velocities, namely $U_g/U_{t,40\mu m}$ values of 1.01 (Fig. 2.15a) and 2 (Fig. 2.15b).

Table 2.6. Base case parameters for binary size Geldart Group A particles.

Parameter	Symbol	Unit	Magnitude
Solid (FCC)	d_{pi}	μm	40 / 75
	ρ_p	kg/m^3	1450
	Φ	-	0.58
	ε_{mf}	-	0.42
	U_t	m/s	0.037 / 0.11
Air	ρ_g	kg/m^3	1.2
	μ_g	kg/m/s	1.8×10^{-5}
Fluidizing unit	D_c	m	0.5
General	T	K	298
	G	kg/m/s^2	9.81

With regards to the maximum orders of magnitude difference among the correlations, the range of 5 to 13 is observed for the binary mixtures at both $U_g/U_{t,40\mu\text{m}}$ values (Figs. 2.15a and b). Generally, at the specified $U_g/U_{t,40\mu\text{m}}$, K_∞ increases with mass fraction of the finer FCC, which is reasonable since the driving force experienced by 100 % finer FCC (at x -axis value of 1) is higher than that by 100 % coarser FCC (at x -axis value of 0), and agrees with some experimental data [14, 15, 78, 79]. On the other hand, a handful of correlations instead predict that K_∞ remains constant or decreases with mass fraction of finer FCC, which also corroborates with experimental data specifically with Group A or C as the finer species [80-82], on the basis that inter-particle cohesion interactions increasingly diminishes K_∞ as composition of fines increases. Specifically, one correlation [37] indicate that K_∞ remains constant at both

the lower $U_g/U_{t,40\mu m}$ (Fig. 2.15a) and higher $U_g/U_{t,40\mu m}$ (Fig. 2.15b), primarily because the correlation only accounts for U_g and disregards particle properties entirely. With regards to correlations predicting that K_∞ decreases with mass fraction of finer FCC, one [49] did so both at the lower $U_g/U_{t,40\mu m}$ (Fig. 2.15a) and higher $U_g/U_{t,40\mu m}$ (Fig. 2.15b), respectively. This sole correlation [49] that predict decreasing K_∞ for both $U_g/U_{t,40\mu m}$ values did so based on experimental results suggesting that the K_∞ equation be tweaked for three different ranges of d_{pi} values of bed particles (namely for $d_{pi} \leq 60 \mu m$, $60 \mu m \leq d_{pi} \leq 200 \mu m$, $d_{pi} \geq 200 \mu m$). More specifically, the counter-intuitive trend depicted here is because the two species in this case traverse one of the size demarcations, since subsequent plots show that predicted K_∞ increases with mass fraction of the finer or less dense species (Figs. 2.15 – 2.16).

Fig. 2.15. Total entrainment flux (K_∞) for binary size FCC particles (Group A) as a function of mass fraction of finer FCC at $U_g/U_{t,40\mu m} =$ (a) 1.01 and (b) 2.00.

Analogous to Table 2.6, Table 2.7 lists the key parameters for the binary density Geldart Group A mixture, whose constituent particles are FCC and glass beads of the same size ($d_{pi} = 40 \mu\text{m}$). Notably, the ratios of the U_t values between the two species are the same for both the size and density binary mixtures.

Table 2.7. Base case parameters for binary density Geldart Group A particles.

Parameter	Symbol	Unit	Magnitude
Solid (FCC, glass)	d_{pi}	μm	40
	ρ_p	kg/m^3	1450 / 2500
	Φ	-	0.58 / 0.95
	ε_{mf}	-	0.42
	U_t	m/s	0.037 / 0.11
Air	ρ_g	kg/m^3	1.2
	μ_g	kg/m/s	1.8×10^{-5}
Fluidizing unit	D_c	m	0.5
General	T	K	298
	G	kg/m/s^2	9.81

For this binary mixture, Fig. 2.16 shows that two correlations [36, 37], instead of one [37] for the size binary mixture (Fig. 2.15), predict that K_∞ is invariant with changes in composition of the binary mixture. In this case, the only particle property that Lin et al. [36] took into account is size (d_{pi}), and hence K_∞ erroneously disregards the density difference between the two species in the binary mixture. On the other hand, one [68] and nine [1, 27, 28, 38, 57, 58, 63, 68, 69] correlations predict the unphysical phenomenon that K_∞ decreases with mass fraction of FCC (species with lower U_t) at the

lower $U_g/U_{t,FCC}$ (Fig. 2.16a) and higher $U_g/U_{t,FCC}$ (Fig. 2.16b), respectively. Apparently, extrapolation of existing empirical correlations, even those developed for multiple species, to binary mixtures does not work well, as further evidenced in the 6 to 13 orders of magnitude difference among the correlations.

Fig. 2.16. Total entrainment flux (K_{∞}) for binary density FCC and glass particles (Group A) as a function of mass fraction of FCC at $U_g/U_{t,FCC} =$ (a) 1.01 and (b) 2.00.

• Geldart Group B particles

Analogous to Tables 2.6 and 2.7, the base case parameters for binary Group B mixtures are given in Table 2.8 and Table 2.9 for the size and density systems, respectively. Similarly, for a fairer comparison between the two binary mixtures, the ratios of the U_{ti} values are the same at 2.2. In the case of the size binary mixture (Table 2.9), the two-constituent species are glass beads of d_{pi} values of 225 and 500 μm . On

the other hand, in the case of the density binary mixture (Table 2.9), the two-constituent species are polystyrene (PS) and glass beads with the same d_{pi} of 500 μm .

Table 2.8. Base case parameters for binary size Geldart Group B particles.

Parameter	Symbol	Unit	Magnitude
Solid (glass)	d_{pi}	μm	225 / 500
	ρ_p	kg/m^3	2500
	Φ	-	0.95
	ε_{mf}	-	0.42
	U_t	m/s	1.7 / 3.8
Air	ρ_g	kg/m^3	1.2
	μ_g	kg/m/s	1.8×10^{-5}
Fluidizing unit	D_c	m	0.5
General	T	K	298
	G	kg/m/s^2	9.81

Along the same vein, Fig. 2.17 and Fig. 2.18 display the predicted K_∞ values for the size and density binary mixtures, respectively, as functions of the species with the higher U_t at the U_g/U_t values of 1.01 and 2.00. Generally, K_∞ increases with mass fraction of the species with lower U_t , agreeing with experimental data whereby the species with the lower U_t is either Group A or B [14, 15, 78, 79]. Thereby, in contrast to the previous discussion on Group A wherein inter-particle cohesive interaction may be more prominent, experimental results indicate that K_∞ should increase as composition of the species with lower U_t increases for Group B particles [15]. However, for the binary size mixture (Fig. 2.17), two [37, 49] and four [37, 49, 59, 60] correlations predict

that K_{∞} either remains constant or decreases as the composition the smaller species increases at the lower $U_g/U_{t,225\mu m}$ (Fig. 2.17a) and higher $U_g/U_{t,225\mu m}$ (Fig. 2.17b) values, respectively. Furthermore, for the binary density mixture, six [36, 37, 59, 67-69] and thirteen [1, 13, 27, 36-38, 49, 58-60, 63, 68, 69] correlations predict that K_{∞} either remains constant or decreases as the composition of the less dense PS species increases at the lower $U_g/U_{t,PS}$ (Fig. 2.18a) and higher $U_g/U_{t,PS}$ (Fig. 2.18b) values, respectively. Regarding discrepancies among the correlations, the orders of magnitude difference between the minimum and maximum K_{∞} values are about 4 to 17 for all four graphs in Fig. 2.17 and Fig. 2.18, which again highlights the huge discrepancies among existing empirical correlations in prediction of K_{∞} values beyond the tested experimental parameters.

Table 2.9. Base case parameters for binary density Geldart Group B particles.

Parameter	Symbol	Unit	Magnitude
Solid (PS, glass)	d_{pi}	μm	500
	ρ_p	kg/m^3	1050 / 2500
	Φ	-	0.80 / 0.95
	ε_{mf}	-	0.42
	U_t	m/s	1.7 / 3.8
Air	ρ_g	kg/m^3	1.2
	μ_g	$kg/m/s$	1.8×10^{-5}
Fluidizing unit	D_c	m	0.5
General	T	K	298
	G	$kg/m/s^2$	9.81

Fig. 2.17. Total entrainment flux (K_∞) for binary size glass particles (Group B) as a function of mass fraction of finer glass at $U_g/U_{t,500\mu\text{m}} = (a) 1.01$ and $(b) 2.00$.

Fig. 2.18. Total entrainment flux (K_∞) for binary density PS-glass particles (Group B) as a function of mass fraction of PS at $U_g/U_{t,PS} = (a) 1.01$ and $(b) 2.00$.

2.3. Conclusions

This effort highlights the inadequacies of available entrainment correlations through a comprehensive review and corresponding application to polydisperse, monodisperse, and binary systems of Geldart Groups A and B particles. A few exceptional observations warrant reiteration here: (i) the discrepancies in predicted K_{∞} values are up to 20 orders of magnitude, and (ii) the unphysical preferential elutriation of larger particles is predicted by some correlations. Collectively, the general lack of predictive capability of correlations in applicability beyond the specific operating conditions, experimental setup, and particle used to develop the correlation is demonstrated, hence highlighting the limited usefulness of each correlation. As apparent, the *ad hoc* inclusion or exclusion of parameters or dimensionless groups to improve empirical data-fitting is not effective in generating a comprehensive entrainment correlation. In particular, the lack of understanding of inter-particle effects, namely cohesive agglomeration of smaller particles, dynamic clustering due to hydrodynamic instabilities, and inter-species collisional momentum transfer, seem largely culpable for the huge discrepancies in the predicted K_{∞} values. Therefore, the review suggests the need for more physically-based models based on fundamental descriptions to enable more accurate quantitative predictions of the elutriation and entrainment phenomena.

Nomenclature

A	= cross-sectional area of fluidizing column, m^2
Ar	= $\frac{gd_{pi}^3\rho_g(\rho_p-\rho_g)}{\mu^2}$ = Archimedes number of size i particles, –
C_d	= drag coefficient, –
D_c	= inner diameter of fluidizing column, m
d_{pi}	= mean particle size of size i particles, m
d_b	= equivalent bubble size at bed surface, m
Fr_i	= $\frac{(U_g-U_{ti})^2}{gd_{pi}}$ = particle Froude number of size i particles, –

g	= gravitational acceleration, m/s ²
H	= freeboard height, m
H_c	= height of fluidized bed column, m
H_s	= height of settled fluidized bed, m
$K_{i\infty}$	= elutriation rate constant, kg/m ² s
Re_p	= $\frac{U_g d_{pi} \rho_g}{\mu}$ = particle Reynolds number evaluated at U_g , –
Re_i	= $\frac{(U_g - U_{ti}) d_{pi} \rho_g}{\mu}$ = particle Reynolds number evaluated at $U_g - U_{ti}$, –
Re_t	= $\frac{U_{ti} d_{pi} \rho_g}{\mu}$ = particle Reynolds number evaluated at U_{ti} , –
Re_D	= $\frac{U_g D_{col} \rho_g}{\mu}$ = flow Reynolds number, –
T	= temperature, K
U_g	= superficial gas velocity, m/s
U_{mfi}	= minimum fluidization velocity of size d_{pi} particles, m/s
U_{ti}	= terminal velocity of size d_{pi} particles, m/s
x_i	= mass fraction of size d_{pi} , –
x_{ie}	= mass fraction of size d_{pi} in elutriated material, –

Symbols and Greek Letters

ε_{mf}	= voidage under minimum fluidization condition, –
μ_g	= gas viscosity, kg/m/s
ρ_g	= gas density, kg/m ³
ρ_p	= particle density, kg/m ³
ϕ	= particle sphericity, –

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CHAPTER 3: PREDICTION OF TRANSPORT DISENGAGING HEIGHT (TDH) FOR MONODISPERSE AND POLYDISPERSE SYSTEMS

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Abstract

Transport Disengaging Height (TDH), defined as the freeboard height whereby the entrainment rate does not change appreciably [1-6], is an important parameter in the design of gas-solid fluidized bed systems to minimize particle loss. Unfortunately, despite the initiation and subsequent development of TDH correlations since 1958 [4], poor agreement between predicted and experimental values persists [1] due to the reliance on empirical data-fitting in the absence of a fundamental understanding of the TDH phenomenon. Accordingly, this work aims to provide a comprehensive review of the available TDH correlations.

TDH values of various polydisperse systems predicted by 25 correlations were evaluated over a range of superficial gas velocities, particle sizes, and column diameters. Four observations are worth highlighting: (i) Discrepancies of up to five orders of magnitude were found among TDH values predicted by the various correlations, (ii) Unphysical phenomena predicted include negative TDH values, (iii) Geldart Group B

correlations perform better for Geldart Group B particles than Geldart Group A correlations for Geldart Group A particles, (iv) Prediction often fails in the transition from the freely bubbling to the slugging regime.

The *ad hoc* inclusion and/or exclusion of parameters, and the use of empirical constants to improve empirical data-fitting in the generation of empirical TDH correlations are not useful in either improving predictions of TDH values or advancing the understanding of the TDH phenomenon. Correlations empirically derived do not perform well beyond the narrow scope of experimental condition tested, while semi-empirical or theoretical models available fall short. The lack of predictive capability of the available TDH correlations appears to stem from a deficiency in an understanding of the impact of inter-particle (e.g., cohesion or clustering effects) and inter-species interactions (e.g., collisional momentum transfer effects).

Keywords: transport disengaging height; gas-solid fluidization; Geldart Group A; Geldart Group B; monodisperse, binary mixture, continuous particle size distribution

3.1. Introduction

Gas-solid fluidized bed technology finds widespread industrial applications due to the various advantages offered, including uniform temperature distribution, and excellent mass and heat transfer. Unfortunately, the predictive capability of such processes is still hampered by the lack of understanding related to the complex flow dynamics and inter-particle interactions in fluidized bed systems [7-10].

Transport Disengaging Height (TDH), defined as the freeboard height beyond which the entrainment rate or particle concentration does not change appreciably [1-6], is an important parameter to ensure efficient and sustainable cyclone performance.

Physically, TDH is associated with entrainment, which is generally acknowledged as being initiated by bubble eruption at the bed surface [11, 12], which in turn ejects both fine (i.e., particles whose terminal velocity U_{ti} is less than the operating superficial gas velocity U_g) and coarse particles (whereby $U_{ti} > U_g$) from the dense bed into the freeboard [11, 13, 14]. As particles travel upwards in the freeboard, some particles will return to the dense bed while others will continue the upward trajectory, thereby resulting in an exponentially decreasing entrainment or particle concentration profile with respect to height [2, 5, 15-19]. TDH is acknowledged to be affected by several factors, such as particle properties (i.e., size, density), gas properties (i.e., density, viscosity), superficial gas velocity, bubble characteristics (i.e., diameter, velocity), column dimension (i.e., diameter, height), internals (i.e., tubes, baffles), and operating conditions (i.e., temperature, pressure) [5, 11, 20-31]. Unfortunately, despite the progressive development of TDH correlations since 1958 [4], poor agreement between predicted and experimental values persists, and huge discrepancies exist among the various correlations. Two hypotheses are put forth to underlie the discrepancies.

First, the available TDH correlations (Table 3.1) are largely derived from curve-fitting of experimental data, so it is worthwhile noting that the measurement of TDH values experimentally deviates on two key aspects: (i) the different definitions of TDH, and (ii) the different measuring instruments used. With regards to (i), although TDH is generally defined as the height whereby entrainment or particle concentration becomes relatively constant with respect to height [1-6], the variation of this definition is embodied in the inconsistencies in pinpointing the onset of constancy with height. Specifically, authors have defined TDH as the height whereby (a) particle velocity in the freeboard equals to superficial gas velocity [4, 23], (b) the jet velocity is less than 1 % [32] or 5 % [33, 34] of its magnitude at the bed surface, (c) the entrainment flux is

less than 1 % [27, 35, 36], 3% [37, 38], 4% [4, 39], 5 % [40], or 6 % [21, 41] of the flux at the bed surface, (d) the change in entrainment rate with height is less than 1 % [16], (e) the entrainment flux is 0.1 g/m²/s [32] or 3 g/m²/s [42], or (f) bubble fraction in the gas phase is 0.5 [28]. Such differences in the experimental identification of TDH inevitably lead to mismatched results. As for (ii), the instruments used include video imaging [43], hot wire anemometer [5, 34], fiber optic probe [34, 44], isokinetic sampling probe [5, 36, 40, 45], pressure gauge [11, 22, 23, 27-30, 42, 45-48], Laser Doppler Velocimetry (LDV) [33], and Positron Emission Particle Tracking (PEPT) [49]. These instrumentations differ not only in the characteristic measured but also confer different sensitivity, hence it is not surprising that the various independent instruments lead to disparate TDH results.

Second, another source of discrepancy lies in the twin definitions of TDH, namely TDH_C and TDH_F [28, 29]. While TDH_C or splash height is the maximum height attained by coarse particles ($U_{ti} > U_g$) [28, 29, 34, 50], TDH_F is the commonly known TDH, i.e., the height where the change of particle concentration in the freeboard is negligible [4, 5]. TDH_C applies specifically for the coarser Geldart Groups B and D particles while TDH_F for the finer Geldart Groups A and C particles [1, 29], but the TDH generalized for all Geldart Groups typically refers to TDH_F. Nonetheless, it is important to note that the two definitions of TDH would result in differences in the TDH values reported, especially since TDH_F is higher than TDH_C at higher gas velocity, and vice versa at lower gas velocity [49].

Modeling efforts have also progressed in parallel with the experiments, with Brems et al. [49] recently developing a purely theoretical TDH correlation from force balance. The other semi-empirical TDH correlations available largely stem from two core concepts, namely, (i) jet dissipation theory accounting for hydrodynamics

instability due to bubble eruption at the bed surface, and (ii) force balance of particles (either as single particles or clusters) in the freeboard. Zenz and Weil [4], George and Grace [23], Horio et al. [34], and Pemberton and Davidson [36] were among the first researchers who developed such semi-empirical TDH correlations, whereby TDH was defined as the height at which the jet velocity is equal to the superficial gas velocity. Of these, only Horio et al. [34] included the effect of turbulence. Subsequently, Do et al. [43] modified the Zenz and Weil correlation [4] by changing the simplified drag force (for laminar flow) with the standard drag force [51] (for both laminar and turbulent flow), while Fung and Hamdullahpur [32] used jet dissipation theory (hitherto only used for fine particles) to formulate the TDH correlation of large particles ($U_{ti} > U_g$) without the presence of fine particles ($U_{ti} < U_g$). Regarding (i), the concept of ‘ghost bubble’ (i.e., imaginary bubble in the freeboard with similar identity as the gas originating from bubble eruption at the bed surface [52]), was postulated by Pemberton and Davidson [52] to be an improvement of the jet dissipation theory, which does not account for gas turbulence in the freeboard following bubble eruption at the bed surface. In this case, TDH is defined as the height at which the ‘gas puff’ originating from the bubble burst at bed surface is dispersed into the bulk gas flow. Following this, Hamdullahpur and Mackay [33], Sciazko et al. (1991) [29], and Fung and Hamdullahpur [32] further developed the ‘ghost bubble’ concept. With respect to (ii), Baron et al. [11, 48] and Kim et al. [42] were among the first authors who assumed that particles are ejected from the bed surface as clusters instead of individual particles, which has direct implications on the force balance of the particles in the freeboard. The non-negligible influence of clusters has motivated other researchers to find new drag correlations.

The objective of the current review is evaluating the applicability of available TDH correlations to common Geldart Groups A and B test cases and thereby identifying

the gaps in the knowledge base underlying the discrepancies among the correlations. Two notes are worth highlighting: (1) New experimental data is not reported here, since the generation of another empirical TDH correlation is anti-thesis to the gist of the review; and (2) The evaluation is not a comparison of measured and predicted values, but a comparative analysis of the TDH values predicted by the correlations. Specifically, the aim of the current effort is at understanding the deficiency of existing correlations, thereby provide physical insights for better predictive capability of the TDH phenomenon. In the absence of a more fundamental understanding of TDH, the *ad hoc* use of empirical constants and non-physically-based terms to improve the data-fitting of experimental data appear to restrict the utility of such correlations only to the narrow scope tested. In addition, the semi-empirical or theoretical models available also fall short due to a gap in the knowledge base. Four highlights emanate from this review: (1) Discrepancies in TDH values predicted are up to five orders of magnitude and contradictory trends exist among the 25 correlations available; (2) Unphysical TDH trends are predicted, namely, (i) negative TDH for fine particles ($U_{ti} < U_g$) [43] and slugging conditions [28], (ii) TDH decreases [28, 47] or remains constant as U_g increases, especially when the system transitions into the slugging regime [11, 23, 32-34, 36, 44, 48], and (iii) TDH is constant with column diameter (D_c) for $D_c < 0.6$ m [5, 27, 29, 35, 40, 45, 46], although the dominant wall effect for increasingly smaller columns is expected to affect TDH [16]; (3) TDH correlations developed based on Geldart Group B particles show better agreement compared to those based on Geldart Group A, presumably due to the augmented inter-particle cohesive effects in the latter; (4) The understanding of the TDH phenomena is lacking for the slugging regime relative to freely-bubbling regime.

Table 3.1. TDH correlations.

Year	Author(s)	Correlations	Geldart Group	Particle	ρ_p (kg/m ³)	d_{pi} (μ m)	U_g (m/s)	D_c (m)
1958	Zenz and Weil. [4]	$TDH = \frac{\rho_p d_{pi}^2}{28.1 \mu_g} \left[U_{psi} + U_{mi} \ln \left(1 - \frac{U_{psi}}{U_{mi}} \right) \right]$ $U_{mi} = U_g - \frac{1.15 \rho_p d_{pi}^2}{\mu_g}$	A	FCC catalyst			0.3- 0.72	2D column (0.051x 0.61)
1968	Amitin et al. [40]	Based on solid concentration: $TDH = 0.85 U_g^{1.2} (7.33 - 1.2 \log_{10} U_g)$ Based on particle trajectory: $TDH = \frac{3.2 \sqrt{D_c}}{U_{ti} - U_g}$	C, A, B	Catalyst	1400	1-32	0.2- 0.6	0.144
1969	Soroko et al. [46]	Without stabilizing grid: $TDH = 1200 H_s Re_{pi}^{1.55} Ar_i^{-1.1}$	B, D	Catalyst		750- 2500		3; 3.5

		With stabilizing grid: $TDH = 730H_s Re_{pi}^{1.45} Ar_i^{-1.1}$						
1972	Do et al. [43]	$TDH = \frac{\rho_p d_{pi}^2 U_{psi}^2}{28.125 \mu_g} \left[1 - \frac{1}{V} \ln(1 + V) \right]$ $V = \frac{U_{psi}}{U_{ti} - U_g}$	B	glass beads		177- 250	5.4- 10.8	(2D column)
1972	Frantz and Juhl [22]	$TDH = 8.4 \times 10^4 d_{pi} \rho_g^{0.33} \mu_g^{0.29} D_c^{0.16} U_g^m$ $m = D_c^{-0.13}$ *TDH, d_{pi} , and D_c in ft; ρ_g in lb/ft ³ ; μ_g in lb/ft/hr; U_g in ft/s	A, B	sand, catalyst	818- 4072	46- 305	0.000 24 - 0.67	0.025- 0.152
1973	Fournol et al. [5]	$TDH = 1000 \frac{U_g^2}{g}$	A	FCC catalyst	830	58	0.11- 0.22	0.61
1978	George and Grace [23]	$TDH = 18.2 d_{bi}$	A, B, D	glass, coke, FCC catalyst, silica sand	1900, 2640	20- 500	0.4- 9.3	0.108, 0.457

1978	Tanaka and Sinhara [35]	$TDH = \ln\left(\frac{100}{a_c}\right)$ $a_c = 2.34 \times 10^{-5} \frac{\rho_p U_g (U_{ti}/U_g)^{-0.7}}{Fr_i \mu_g}$	D	Sand	2670	718	1.8- 2.4	
1980	Horio et al. [34]	$TDH = \frac{112}{\eta} u'_{vo} \left(\frac{d_{bs}}{g}\right)^{0.5}$ $\eta = 4.5; u'_{vo} = 0.565$	B	glass beads		164- 1350	0.3- 0.6	0.24
1982	Wen and Chen [16]	$TDH = \left(\frac{1}{a_c}\right) \ln\left(\frac{K_0 - K_\infty}{0.01 K_\infty}\right)$ K_0 $= 3.07 \times 10^{-9} \frac{\rho_g^{3.5} g^{0.5} (U_g - U_{mf})^{2.5} A d_{bs}}{\mu_g^{3.5}}$	A, B, D		860- 7850	37- 3400	0.1-10	
1983	Hatano and Ishida [44]	$TDH = \frac{U_{psi}^2}{2g}$	A, B	FCC catalyst, glass beads		75, 180	0.15- 0.40	0.15

1983	Horio et al. [85]	$TDH = (2.7D_c^{-0.36} - 0.7)D_c \exp(0.74U_g D_c^{-0.23})$	A					
1984	Chan and Knowlton [27]	$TDH = 0.85U_g^{1.2}(7.33 - 1.2 \log U_g)$	A, B	Sand	2595	37-420		0.3
1986	Pemberton and Davidson [36]	$TDH = \frac{4.605}{\beta}$ $\beta = \frac{1}{3d_{bi}}$	A, B, D	Sand		73-760		0.6
1986	Hamdullahpur and MacKay [33]	$TDH = 11.7 \frac{d_{bi}}{g}$	B	Sand	2600	300	0.2-0.4	0.319 x 0.176 (rectangular)
1986	Yoon et al. [47]	$TDH = 1.38D_c Ar_i^{0.42} Fr^{-0.07}$	A	Coal	2000	34-120	0.3-1.37	0.145

1987	Baron et al. [48]	$\text{TDH} = \frac{(2.1U_{bi})^2}{2g}$ <p>or, $\text{TDH} = \left(\frac{1}{a_{cf}}\right) \ln(K_0 x 10^4)$</p>	A	Sand	2650	75	0.15- 0.30	0.61
1988	Sciazko et al. [28]	$\text{TDH} = 3.1d_{bi} \ln\left(\frac{U_{bi} - U_g}{U_g - 2U_{mf}}\right)$	A, B, D	Char	1262	80- 2000	0.51- 1.23	0.3
1988	Baron et al. [11]	$\text{TDH} = \frac{U_{ci}^2}{2g}$ <p>$U_{ci} = 2U_{bi}$ (for bubbling regime)</p> <p>$U_{ci} = 7.5U_{bi}$ (for slugging regime)</p>	A, B	silica sand		10- 203		0.61
1991	Sciazko et al. [29]	$\text{TDH} = \frac{1500H_c Re_{pi}}{Ar_i}$	A, B, D	Char	1250	80- 2000	0.51- 1.23	0.3
1993	Fung and Hamdullahpur [32]	$\text{TDH} = -3d_{bi} \ln 0.01$	B	Sand		200- 427	0.27- 0.54	0.145

1996	Kim et al. [42]	$TDH = \left(\frac{1}{a_c}\right) \ln\left(\frac{K_0}{0.003}\right)$	A, B, D	PVC, sand	1400- 2620	130- 610	0.32- 0.55	0.3
1997	Smolders and Baeyens [30]	$TDH = 6[(U_g - U_{mf})d_{bs}]^{0.6}$	A, C	limestone, α -alumina, crushed glass, sand, SiC, E-cat, Al_2O_3	1420- 3650	27- 54	0.1- 0.8	0.082
2011	Brems et al. [49]	$\frac{d(TDH)}{dt} = U_g - U_{ri}$ $\frac{d(U_{ri})}{dt} = a + bU_{ri}^2$ for $U_{ri} < 0$ $\frac{d(U_{ri})}{dt} = a - bU_{ri}^2$ for $U_{ri} > 0$ $a = \frac{g(\rho_p - \rho_g)}{\rho_p}$; $b = \frac{3\rho_g C_d}{4\rho_p d_{pi}}$	A, B	sand, alumina, coal	2260, 1600, 1600	150- 550, 135, 390	0.029- 0.352	
2012	Ko [45]	$TDH = 5.66 \left[\frac{Fr_i \rho_g}{Re_{pi}(\rho_p - \rho_g)} \right]^{0.23}$	A, B, D	iron ore	3980	63- 1000	0.36- 2.93	0.11

Table 3.2. Equations for calculating the common parameters in TDH correlations.

Parameter	Equation	Source
Particle drag coefficient	$C_d = \frac{24}{Re_{pi}} \left[1 + 8.1716e^{-4.0655\phi} Re_{pi}^{0.0964+0.5565\phi} + \frac{73.69 Re_{pi} e^{-5.0748\phi}}{Re_{pi} + 5.378e^{6.2122\phi}} \right]$	Haider and Levenspiel (1989) [113]
Bubble diameter	<p>For porous plate: $d_{b0} = 0.376(U_g - U_{mf})^2$</p> $d_{bm} = 1.49[D_c^2(U_g - U_{mf})]^{0.4}$ $d_{bi} = d_{bm} - (d_{bm} - d_{b0}) \exp\left(-0.3 \frac{H_d}{D_c}\right)$	Mori and Wen (1975) [114]
Bubble diameter at bed surface	$d_{bs} = 0.474 \left(\frac{U_g - U_{mf}}{\alpha \sqrt{g}} \right)^{0.4} (H_c + 3.94 \sqrt{A_0})^{0.8}$	Wen and Chen (1982) [16]
Bubble velocity	$U_{bi} = U_g - U_{mf} + 0.71 \sqrt{g d_{bi}}$	Davidson and Harrison (1963) [115]
Velocity of single isolated slug	$U_{sl\infty} = 0.35 \sqrt{g D_c}$	Hovmand and Davidson [116]
Particle velocity at bed surface	$U_{psi} = 2U_{bi}$	Pemberton and Davidson (1986) [13]

3.2. Results and Discussion

Assessment of the available TDH correlations (Table 3.1) through application to continuous PSDs, monodisperse particles, and binary mixtures of Geldart Groups A and B particles, as well as the effect of column diameter will be carried out. To provide an overview of the base case parameters evaluated, Tables 3.3 and 3.4 summarize the particle properties and test conditions appraised in the following sections.

Fig. 3.1 shows predicted TDH values of continuous PSDs Geldart Group A particles (see Table 3.3) with PSD width (σ/d_{sm}) of 0.25 at various $U_g/U_{ti,80\mu m}$. Comparison between Fig. 3.1a (using all available TDH correlations in Table 3.1) and Fig 3.1b (using only TDH correlations developed exclusively for Geldart Group A system) becomes base of the following three exceptions which will be applied throughout current study. First, two correlations in Table 3.1 [16, 42] are omitted from all analyses due to the presence of the entrainment flux parameters (K_0 and K_∞), which have been shown to display discrepancies of up to 20 orders of magnitude among values predicted by available correlations [54]. Second, because the abundance of correlations hampers readability of the figures (see Fig. 3.1a), in the ensuing sections, correlations that superimpose on one another are largely omitted from the figures and represented by a couple of representative plots. Third, application of the TDH correlations (Table 3.1) is not restricted specifically to the Geldart Group from which they were developed, in order to highlight the observation that such a limitation does little to diminish the discrepancies (see Fig. 3.1a and Fig. 3.1b).

Table 3.3. Base case parameters for Geldart Group A particles (TDH study).

Parameters	Symbol	Unit	Continuous PSD	Monodisperse	Binary Size	Binary Density
Particle	-	-	FCC	FCC	FCC	FCC / glass beads
	d_{sm}	μm	80	25 – 200	40 / 75	40
	ρ_p	kg/m^3	1450	1450	1450	1450 / 2500
	ϕ	-	0.58	0.58	0.58	0.58 / 0.95
	ε_{mf}	-	0.42			
	U_{ti}	m/s	0.20	0.015 - 0.48	0.037 / 0.11	0.037 / 0.11
Gas (air)	ρ_g	kg/m^3	1.2			
	μ_g	kg/m/s	1.8×10^{-5}			
Fluidized bed	D_c	M	1			
	H_s	M	1			
Operating condition	g	kg/m/s^2	9.81

Fig. 3.1. Predicted TDH values for lognormal PSDs of Geldart Group A particles (with PSD width expressed as a ratio of standard deviation to Sauter-mean diameter, $\sigma/d_{sm} = 0.25$) as a function of $U_g/U_{ti,80\mu m}$ for: (a) All available TDH correlations, and (b) TDH correlations developed for Geldart Group A.

3.2.1. Continuous PSDs particles

- Geldart Group A particles**

This section applies and evaluates the available TDH correlations (see Table 3.1) for continuous PSDs of Geldart Group A particles [53]. Specifically, the continuous PSDs are mass-based lognormal frequency distributions, as described in a companion review on entrainment [54]. Among the various lognormal PSDs evaluated, the

physically-sound Sauter-mean diameter (d_{sm}) is kept constant, while the widths of the continuous PSDs, defined as the ratio of standard deviation of the PSD to Sauter-mean diameter (σ/d_{sm}), are varied [54]. Table 3.3 lists the base case parameters of the fluidized bed system and the properties of continuous PSD of Fluid Catalytic Cracking (FCC) catalyst, which is the most prevalent Geldart Group A particles in industry [55].

The available TDH correlations are evaluated using lognormal PSDs of various PSD widths (σ/d_{sm}) at various ratios of superficial gas velocity to particle terminal velocity ($U_g/U_{ti,80\mu m}$). While the range of PSD widths illustrate the impact of varying extents of polydispersity, the spectrum of $U_g/U_{ti,80\mu m}$ values depict the impact of varying the driving force for entraining the particles.

Fig. 3.2a and Fig. 3.2b show the TDH values predicted by the available correlations for $U_g/U_{ti,80\mu m}$ values in the range of 1.01 – 2.00 at the narrowest continuous PSD width ($\sigma/d_{sm} = 0.25$) and widest PSD width ($\sigma/d_{sm} = 0.70$), respectively. The velocity range considered is such that particles in the fluidized bed column are incipiently entrained at $U_g/U_{ti,80\mu m} = 1.01$ and substantial entrainment occurs beyond $U_g/U_{ti,80\mu m} = 2.00$ [54], which approximately denotes the transition to the fast fluidization regime [17]. Notably, although measurement of TDH is typically carried out for the bubbling and turbulent regimes, TDH has also been evaluated for the slugging regime [11, 39, 56].

Fig. 3.2. Predicted TDH values for lognormal PSDs of Geldart Group A particles as a function of $U_g/U_{ti,80\mu m}$ for $\sigma/d_{sm} =$ (a) 0.25 and (b) 0.70.

Fig. 3.2 shows expectedly that TDH generally increases with $U_g/U_{ti,80\mu m}$ [4, 5, 11, 27, 40, 43, 49, 50]; higher U_g increases TDH because (i) the number of particles ejected from the bed and hence particle concentration in the freeboard increases, and (ii) the rate of decay of particle concentration in the freeboard decreases [15]. Exceptions are found in Yoon et al. [47] and Sciazko et al. (1988) [28], whose predicted unphysical trend of TDH decreasing with $U_g/U_{ti,80\mu m}$ increasing stem from the negative sign [28] or negative power [47] of U_g in the correlations. The increase of predicted TDH values as $U_g/U_{ti,80\mu m}$ increases from 1.01 to 2.00 varies among the correlations, with the largest

being up to one order of magnitude [5, 46]. Notably, Do et al. [43] is excluded from Fig. 3.2 because negative TDH values are predicted. The negative values are rooted in the $(U_{ti} - U_g)$ term, which bars application to the finer particles with $U_{ti} < U_g$ in a polydisperse system. The $(U_{ti} - U_g)$ term arose because the Do et al. correlation [43] was developed experimentally (in two-dimensional column) using larger Geldart Group B particles at $U_{ti} > U_g$.

Discrepancies of up to four orders of magnitude among predicted TDH values are observed in Fig. 3.2. The huge discrepancies underscore the limited usefulness of the available correlations in predicting TDH values beyond the scope studied. TDH values predicted are largely within an order of magnitude, but outlying high TDH values are predicted by Soroko et al. (both with and without stabilizing grid) [46], Fournol et al. [5], Sciazko et al. (1991) [29], while outlying low TDH values are predicted by Zenz and Weil [4] and Brems et al. [49]. Notably, being an outlier or not has no bearings on the accuracy of the prediction, since it is beyond the scope of this review, whose objective is to understand the underlying physics behind the discrepancies through a comparative analysis of the available TDH correlations. Notably though, other than Soroko et al. [46], the other outlying correlations were developed for Geldart Group A particles, whose inter-particle cohesive effects presumably led to worse predictions due to the lack of physical understanding. In particular, for Soroko et al. [46], what stands out is the large fluidizing column diameter of 3 meters, which is a few times larger than the other setups, although whether the root cause lies in the column diameter is not clear. The other outlying high correlation by Sciazko et al. (1991) [29] was obtained from the modification of the empirical constants in Soroko et al. [46] (based on new experimental data on wider particle size distributions (PSDs)), hence also inherited the high TDH predictions. The third outlying high prediction by Fournol et al. [5] stems from a trickle

valve configuration, which re-entrains particles that return to the bed, hence the TDH values obtained experimentally are considerably higher, as acknowledged by the authors.

Regarding the outlying low TDH predictions (Zenz and Weil [4] and Brems et al. [49]), further inspection reveals some interesting physics. With respect to Zenz and Weil [4], two notes are worth highlighting: (i) unlike the other force-balance-based correlations [23, 34, 43, 49] which use the standard drag curve by Turton and Levenspiel [51], it is the only one which uses the Stokes modified drag correlation which gives lower drag force values; (ii) it is a semi-empirical correlation from an experimental set-up in two-dimensional column that was intended for the evaluation of particle entrainment, hence accuracy involved for TDH is compromised [4, 43], since bubble behavior and particularly bubble eruption are hindered [16, 57]. As for Brems et al. [49], it is the only purely theoretical correlation without any empirical constants. Unfortunately, the embedded assumption that the initial particle velocity is equal to bubble velocity at the bed surface is the root cause of lower TDH predictions, because, in practice, the bubble velocity at the bed surface underestimates the actual particle ejection velocity, which has been reported to be around twice that of the bubble velocity at the bed surface [14, 23, 43, 44, 58-60]. Nevertheless, Brems et al. [49] provides a good framework for furthering the development of a theoretical TDH model.

A more in-depth discussion on the force balance in a simple ballistic model, which both the lowest TDH predictions stem from [4, 49], is worthwhile here. Notably, the model is based on the assumptions that the particles move individually and inter-particle interactions (i.e., inter-particle momentum transfer via collision and/or cohesive forces) are negligible. Such a simplified model tends to underestimate the experimental results especially of polydisperse systems, because (i) the use of mean equivalent diameters understate the maximum height achieved by smaller particles [40, 61, 62], (ii)

the extensive interactions among particles in a fluidized bed system cannot be ignored, especially in terms of the cohesive interactions of the Geldart Group A particles considered here, the inter-species momentum transfer in the presence of a wide particle size range PSD particles [23, 26, 63, 64], and clustering effects [65-67]. Regarding clustering, while previous studies confirmed that particles are ejected from the bed as clusters rather than individual particles [1, 44, 68-70], others have observed that particles exist as dynamic clusters in the freeboard [15, 71-74]. Better quantitative prediction of TDH values can be achieved by incorporating cluster drag correlations [36, 75-78] into the models [11, 79, 80], since the lower drag coefficient [81, 82] and higher slip velocity of clusters [83, 84] contribute to higher TDH predictions. Notably, three other correlations [23, 34, 36] based also on the simple ballistic model did not give outlying TDH values per Zenz and Weil [4] and Brems et al. [49], presumably due to 'better' empirical data-fitting constants.

To further understand the predictive capability of the available TDH correlations for continuous PSDs of Geldart Group A particles, Fig. 3.3 shows the predicted TDH values as a function of σ/d_{sm} in the range of 0.1 (approximately monodisperse) and 0.7 (widest PSD considered here) at $U_g/U_{ti,80\mu m}$ values of 1.01 and 2.00. Three noteworthy observations are apparent in Fig. 3.3. First, the discrepancy between the minimum and maximum TDH values predicted spans four orders of magnitude at both the $U_g/U_{ti,80\mu m}$ conditions evaluated. Again, the highest and lowest TDH values predicted are by correlations developed for Geldart Group A particles, namely, Sciazko et al. (1991) [29] and Zenz and Weil [4], respectively. Second, while correlations developed strictly for Geldart Group A particles fail to give a tighter range of TDH predictions even when applied to Geldart Group A system, TDH values based on correlations developed for different Geldart Groups tend to overlap with one another. It can be observed in Fig. 3.3.

that the outlying correlations [4, 29, 49] here are developed from Geldart Group A particles and applied here to a Geldart Group A system; instead, the correlations developed *only* from Geldart Group B [32-34, 46] or Group D [35] particles overlap with and are indistinguishable from the general predicted trends. Clearly, the application of TDH correlations strictly for the Geldart Group from which they were developed does not diminish the discrepancies. Third, although largely consistent trends of TDH values with respect to $U_g/U_{ti,80\mu m}$ are observed in Fig. 3.2, contradictory trends are found among various TDH predictions with respect to PSD width in Fig. 3.3. At the lower $U_g/U_{ti,80\mu m}$ (Fig. 3.3a), three correlations predict a negative correlation between TDH values and PSD width [28, 45, 46], two correlations predict non-monotonic trends (i.e. increases then decreases, or vice versa) [34, 46], and 18 correlations predict a positive correlation between TDH values and PSD width [4, 5, 11, 22, 23, 27, 29, 30, 32, 33, 35, 36, 40, 44, 47-49, 85]. At the higher $U_g/U_{ti,80\mu m}$ (Fig. 3.3a), 11 correlations predict a negative correlation between TDH values and PSD width [23, 28, 29, 33, 34, 36, 44-46, 48], one correlation predict that TDH is relatively independent of PSD width [46], and 11 correlations give a positive correlation between TDH values and PSD width [4, 5, 11, 22, 27, 30, 35, 40, 47, 49, 85]. The contradictory trends presumably are rooted in a lack of understanding of the effect of inter-particle interactions (namely, cohesive forces, inter-particle momentum transfer, and clustering effects), whose impact may vary depending on the experimental set-up and the operating conditions. The decrease of TDH values with PSD width can be attributed to the cluster phenomenon, which has been reported to be more pronounced in a wider PSD of Geldart Group A particles [86, 87]; unfortunately, a fundamental understanding related to the effect of PSD on clustering behavior remains elusive [65, 66, 88, 89].

Fig. 3.3. Predicted TDH values for lognormal PSDs of Geldart Group A particles as a function of σ/d_{sm} for $U_g/U_{ti,80\mu m} =$ (a) 1.01 and (b) 2.00.

Although the impact of superficial gas velocity on TDH is more intuitive and well-understood, the impact of varying widths of continuous PSDs is lesser known [47, 90, 91]. Importantly, this review highlights the importance of providing the PSD data of particles, which regrettably is generally lacking in the literature. PSD width is expected to have a non-negligible effect on TDH, based on previous work elucidating the relationship between PSD and minimum bubbling fluidization, bubble size, particle segregation, and clustering in the freeboard [88, 92, 93]; hence, further study is warranted.

- **Geldart Group B particles**

The base case parameters for continuous PSDs of Geldart Group B particles are displayed in Table 3.4. Similar to the previous section, the lognormal PSDs evaluated here has been described in an earlier companion entrainment review [54].

Table 3.4. Base case parameters for Geldart Group B particles (TDH study).

Parameter	Symbol	Unit	Continuous PSD	Monodisperse	Binary Size	Binary Density
Particle	-	-	glass beads	glass beads	glass beads	polystyrene / glass beads
	d_{sm}	μm	325	80 – 700	225 / 500	500
	ρ_p	kg/m^3	2500	2500	2500	1050 / 2500
	ϕ	-	0.95	0.95	0.95	0.80 / 0.95
	ε_{mf}	-	0.42			
	U_{ti}	m/s	2.58	0.39 – 4.8	1.7 / 3.8	1.7 / 3.8
Gas (air)	ρ_g	kg/m^3	1.2			
	μ_g	kg/m/s	1.8×10^{-5}			
Fluidized bed	D_c	m	1			
	H_s	m	1			
Operating condition	g	kg/m/s^2	9.81			

Fig. 3.4 shows the effect of $U_g/U_{ti,325\mu m}$ on TDH for both the PSD widths (σ/d_{sm}) of 0.25 and 0.70. Similar to Fig. 3.2, discrepancies of up to four orders of magnitude differences exist between the minimum TDH value predicted by Zenz and Weil [4] and the maximum TDH value predicted by Fournol et al. [5] at the highest $U_g/U_{ti,325\mu m}$. Although it can be argued that these two correlations are outlying because they were generated using Geldart Group A particles, note that they similarly give such outlying predictions even when applied to the Geldart Group A particles (Fig. 3.2).

Fig. 3.4. Predicted TDH values for lognormal Geldart Group B particles as a function of $U_g/U_{ti,325\mu m}$ for $\sigma/d_{sm} =$ (a) 0.25 and (b) 0.70.

Unlike for Geldart Group A particles (Fig. 3.2), where the majority of the correlations predict that TDH increases as U_g/U_{ti} increases, the TDH correlations predict contradictory trends of TDH with respect to $U_g/U_{ti,325\mu m}$ for Geldart Group B particles (Fig. 3.4). In the case of $\sigma/d_{sm} = 0.25$ (Fig. 3.4a), eight correlations [11, 23, 32-34, 36, 44, 48] predict that TDH is independent of $U_g/U_{ti,325\mu m}$, one correlation [47] predicts that TDH decreases with $U_g/U_{ti,325\mu m}$, and 13 correlations [4, 5, 22, 27, 29, 30, 35, 40, 45, 46, 49, 85] predict that TDH increases with $U_g/U_{ti,325\mu m}$. Notably, one correlation (Sciazko et al. (1988) [28]) is omitted from Fig. 3.4, because TDH values cannot be calculated in the slugging regime due to the $\ln(U_{bi} - U_g)$ term. Interestingly, the eight correlations [11, 23, 32-34, 36, 44, 48] that predict the invariance of TDH with $U_g/U_{ti,325\mu m}$ for $\sigma/d_{sm} = 0.25$ (Fig. 3.4a) instead predict that TDH increases with $U_g/U_{ti,325\mu m}$ for $\sigma/d_{sm} = 0.70$ (Fig. 3.4b), presumably because these eight correlations depend solely on bubble characteristics (namely, bubble velocity (U_{bi}) or bubble diameter (d_{bi})). The disagreeing trends imply that (i) although bubbles are acknowledged for influencing the movement of particles in the freeboard [94], the consideration of other fluidized bed characteristics on TDH is necessary, and (ii) the dominant factors influencing TDH are different for Geldart Groups A and B.

The underlying reason for the eight correlations [11, 23, 32-34, 36, 44, 48] predicting the unphysical phenomenon that TDH is invariant with $U_g/U_{ti,325\mu m}$ (Fig. 3.4a) relates to the fluidization regime. Specifically, Fig. 3.4a falls into the slugging fluidization regime, while Fig. 3.4b represents both the bubbling and slugging fluidization regimes. Note that the well-acknowledged transition from the bubbling to slugging fluidization regime occurs when bubble diameter is greater than 0.6 of the fluidizing column diameter [6, 17, 95]. In particular for the slugging regime, past work indicated that the slug velocity and slug diameter should be used instead of the

corresponding bubble parameters [6, 17, 95-97], because of the different characteristics between a slug and a bubble, namely, (i) slug velocity is lower than bubble velocity, and (ii) neither slug velocity or slug diameter is a function of U_g . Hence, a constant TDH with respect to U_g is predicted for correlations which are only based on bubble characteristics, as is the case for the eight correlations considered here [11, 23, 32-34, 36, 44, 48].

Analogous to Fig. 3.3, Fig. 3.5 shows the TDH values predicted in the range of PSD width of 0.1 – 0.7 at both lower $U_g/U_{ti,325\mu m}$ (namely, 1.01) and higher $U_g/U_{ti,325\mu m}$ (namely, 2.00). Similar to that for Geldart Group A, huge discrepancies of up to four orders of magnitude are found at the higher $U_g/U_{ti,325\mu m}$ (Fig. 3.5b). Similarly, it is not possible to calculate TDH using the Sciazko et al. (1988) [28] correlation due to the inapplicability of the $\ln(U_{bi} - U_g)$ term in the slugging regime (Fig. 3.5). At both the lower and higher $U_g/U_{ti,325\mu m}$ (Fig. 3.5), as PSD width increases, five correlations [4, 35, 45, 46] predict a decrease of TDH, 13 correlations [11, 23, 27, 29, 30, 32-34, 36, 40, 44, 48] predict non-monotonic trends (i.e., increases then decreases or vice versa), while four correlations [4, 45, 46] predict an increase of TDH. Collectively, the contradictory trends in Fig. 3.3 and Fig. 3.5 highlight the importance of an improved physical understanding of the effect of PSD width for better predictive capability of TDH correlations. Another interesting note is that the Fournol et al. [5] and Zenz and Weil [4] correlations give the highest and lowest predictions, respectively, of TDH values; although both of these correlations were developed for the same Geldart Group (i.e., Geldart Group A), the TDH values predicted are distinctly different. Notably, other researchers [11, 27, 47], including Fournol et al. [5], have indicated that the Fournol et al. [5] correlation generally predicts higher TDH values, due to the use of trickle valve inside fluidized bed column which increases entrainment rate throughout the column

and hence TDH value. Once again, this result indicated that the differences in predicted TDH values arising not from different Geldart Groups which each correlations developed from.

Fig. 3.5. Predicted TDH values for lognormal PSDs of Geldart Group B particles as a function of σ/d_{sm} for $U_g/U_{ti,325\mu m} =$ (a) 1.01 and (b) 2.00.

Comparing the TDH predictions for Geldart Group A (Fig. 3.2 and Fig. 3.3) and Geldart Group B (Fig. 3.4 and Fig. 3.5) systems reveal two interesting observations. First, all of the correlations generally predict higher TDH values for Geldart Group B particles (Fig. 3.4 and Fig. 3.5) relative to Geldart Group A particles (Fig. 3.2 and Fig. 3.3), which stems from the fact that the TDH correlations are largely based on bubble

characteristics (namely, U_{bi} or d_{bi}) or U_g , which values are greater for the larger Geldart Group B particles. It is worthwhile noting that the use of U_g/U_{ti} in the evaluation here (Fig. 3.2, Fig. 3.3, Fig. 3.4, and Fig. 3.5) means that the $(U_g - U_{ti})$ term is greater for the larger Geldart Group B than Geldart Group A particles, which may result in the higher TDH values for Geldart Group B particles. Accordingly, the impact of $(U_g - U_{ti})$ on TDH will be studied in the subsequent sections on monodisperse systems.

Second, the predicted TDH values (sans the obvious outliers) largely fall in a narrower range of approximately one order of magnitude for Geldart Group A (see Fig. 3.2 and Fig. 3.3) compared to the two orders of magnitude for Geldart Group B (see Fig. 3.4 and Fig. 3.5), which is counter to that for entrainment values whereby predictions are worse for Geldart Group A [54]. The underlying reason is likely the lack of understanding related to the slugging regime, which is more prevalent for the Geldart Group B test cases. However, if assessment is restricted to correlations developed only for the same Geldart Group, the TDH values predicted by Geldart Group A correlations for the Geldart Group A test case here (Table 3.3) present discrepancies of four orders of magnitude (Fig. 3.2 and Fig. 3.3), while the TDH values predicted by Geldart Group B correlations for the Geldart Group B test case here (Table 3.4) show a lower discrepancy of two orders of magnitude (Fig. 3.4 and Fig. 3.5). In other words, although the number of TDH correlations intended solely for Geldart Group A [4, 5, 47, 48, 85], is the same as that for Geldart Group B [32-34, 43], the order of magnitude discrepancies among correlations developed for Geldart Group B is lower than that of correlations developed for Geldart Group A.

In summary, based on the observations for the continuous PSDs systems considered here, (i) theoretical [49] or semi-empirical [4] models for TDH are inadequate due to the assumptions used in the absence of understanding rooted in first-

principles, (ii) TDH predictions largely fail in the slugging regime, and (iii) predictive capability of TDH values in the freely bubbling regime is generally better for Geldart Group B than Geldart Group A, presumably due to the lack of understanding of the inter-particle cohesive effects present in the latter.

3.2.2. Monodisperse particles

- **Geldart Group A particles**

Like other fluidization phenomena, TDH is significantly affected by particle properties (i.e., particle size (d_{pi}) and particle density (ρ_p)) [1, 3, 6]. To evaluate the impact of particle size without the inherently complex impact of polydispersity, this section investigates TDH predictions for monodisperse Geldart Group A particles while an analogous assessment is made for monodisperse Geldart Group B particles in the next section. It is important to note that, in practice, a monodisperse system does not refer to one comprising of particles of exactly the same size, but particles with a practically-possible narrow PSD [6]. For evaluation of monodisperse Geldart Group A particles, 80 μm FCC particle is chosen as the test case. Details of the base case parameters used in this section are displayed in Table 3.3.

Previous results on continuous PSDs systems indicate that larger TDH values are obtained for the larger Geldart Group B particles compared to Geldart Group A, because keeping U_g/U_{ti} constant leads to larger $(U_g - U_{ti})$ for larger particles. Hence, in order to illuminate any disparities between the two terms involving U_g , it is expedient to study the predicted TDH values as functions of both $U_g/U_{ti,80\mu\text{m}}$ (dimensionless) and $(U_g - U_{ti,80\mu\text{m}})$ (with unit of m/s), both of which are common terms in entrainment correlations [54]. Fig. 3.6a and Fig. 3.6b portray largely similar trends and magnitudes, which suggests interchangeability between the two embodiments of terms involving U_g .

This section and the next on monodisperse systems serve to provide more insights toward the impact of U_g on TDH for different Geldart Groups (namely, A and B).

Fig. 3.6. Predicted TDH values for monodisperse Geldart Group A particles (FCC) as a function of (a) $U_g/U_{ti,80\mu m}$, and (b) $U_g - U_{ti,80\mu m}$.

By comparing Fig. 3.6a (monodisperse Geldart Group A particles) and Fig. 3.2 (continuous PSDs of Geldart Group A particles), both of which are evaluated as functions of the same range of $U_g/U_{ti,80\mu m}$, two noteworthy observations are apparent. First, monodisperse (Fig. 3.6a) and continuous PSD (Fig. 3.2) systems are similar in that: (i) discrepancies of four orders of magnitude exist between the minimum and maximum TDH values, (ii) the Sciazko et al. (1991) [29] and Zenz and Weil [4] correlations predict

the highest and the lowest TDH values, respectively, and (iii) all of the correlations predict that TDH increases with $U_g/U_{ti,80\mu m}$ or $(U_g - U_{ti,80\mu m})$, except for Yoon et al. [47] and Sciazko et al. (1988) [28]. Regarding (i), that such huge discrepancies persist even for a relatively simple monodisperse system (i.e., in the absence of the effect of polydispersity) in Fig. 3.6 is further evidence that the use of empirical constants and *ad hoc* inclusion of certain terms to data-fit experimental results is inadequate in establishing good predictive capability for TDH values. As for (iii), the terms responsible for the unphysical trends of TDH decreasing with $U_g/U_{ti,80\mu m}$ by Sciazko et al. (1988) [28] and Yoon et al. [47] are the negative orders of non-physically-based terms of U_g and $\ln[(U_b - U_g)/(U_g - 2U_{mf})]$, respectively. Second, TDH values of monodisperse system (Fig. 3.6a) are slightly lower than that of the continuous PSDs system with the same d_{sm} (Fig. 3.2), which presumably stems from inter-species momentum transfer effects, but the understanding of which is lacking.

Fig. 3.7 shows the effect of particle diameter (d_{pi}) at the minimum and maximum U_g/U_{ti} values (namely, 1.01 and 2.00, respectively) considered in this review. Five observations merit attention. First, higher discrepancies of up to five orders of magnitude are observed among the predicted TDH at the lowermost d_{pi} value, presumably due to the lack of a unifying understanding of the inter-particle cohesive interactions and clustering phenomena, which are more dominant for the smallest Geldart Group A particles [30, 98-102]. Second, consistently at both the lower and higher U_g/U_{ti} , eight correlations [11, 23, 32-34, 36, 44, 48] predict that TDH increases steeply at lower d_{pi} values but plateaus off at the higher d_{pi} values, three correlations [28, 46] predict non-monotonic trends (i.e., increases then decreases), and two correlations [29, 45] predict that TDH decreases with d_{pi} . In particular, the eight correlations [11, 23, 32-34, 36, 44, 48] which predict that TDH plateaus off with increasing d_{pi} are those

based on bubble parameters (namely, U_{bi} or d_{bi}); the initial increase of TDH with d_{pi} is because U_{bi} and d_{bi} , increase with d_{pi} in the bubbling regime, whereas the plateauing is due to the independence of slug velocity and size on d_{pi} in the slugging regime. Third, another interesting observation lies in the larger two to three orders of magnitude increase in TDH predicted by seven correlations [4, 5, 11, 22, 27, 40, 49] in the range of d_{pi} considered. Closer inspection reveals a common thread among the correlations, namely, the use of higher (> 1) positive orders of U_g and/or d_{pi} in the correlations. Fourth, distinctive TDH trends are displayed by several correlations at the transition from the bubbling to the slugging regimes. As an example, Baron et al. [11] predicts a distinctly steep increase of TDH with d_{pi} at $d_{pi} = 140 \mu\text{m}$ (Fig. 3.7a) and $d_{pi} = 120 \mu\text{m}$ (Fig. 3.7b), because TDH was assumed to be based on cluster ejection velocity, which was defined differently for the bubbling and slugging regimes (see Table 3.1). As another example, Sciazko et al. (1988) [28] predicts that (i) TDH changes from increasing to decreasing trend at lower U_g/U_{ti} (Fig. 3.7a) and (ii) TDH is negative for higher U_g/U_{ti} (Fig. 3.7b), both of which stem from the non-physically-based $\ln[(U_b - U_g)/(U_g - 2U_{mf})]$ term in their correlations. Fifth, the Soroko et al. [46], Tanaka and Sinohara [35], and Baeyens and Smolders [30] correlations display contradictory trends depending on whether ($U_g - U_{ti}$) or U_g is kept constant; specifically, they predict that TDH increases monotonically [30, 35] or non-monotonically (i.e., increases then decreases) [46] with d_{pi} at constant ($U_g - U_{ti}$), but decreases with d_{pi} at constant U_g . This contradictory trend is due to the opposite signs between U_g and the variables associated with particle size (i.e., d_{pi} , U_{ti} , or U_{mf}). Notably, Sciazko et al. (1991) [29] and Ko [45] also designated U_g and d_{pi} with opposite signs, but contradictory trends are not predicted due to the U_g/d_{pi}^2 term, which decreases the sensitivity of U_g on particle size compared to the previous three correlations [30, 35, 46]. Again, the failure of the available correlations in predicting

TDH beyond the scope experimentally investigated underscores the importance of more physically-based TDH correlations.

Fig. 3.7. Predicted TDH values for monodisperse Geldart Group A particles (FCC) as a function of d_{pi} for $U_g/U_{ti} = (a) 1.01$ and $(b) 2.00$.

• Geldart Group B particles

This section evaluates the TDH predicted by the available correlations for monodisperse Geldart Group B particles, specifically glass beads in the d_{pi} range of 80 - 700 μm . Details of the base case parameters are in Table 3.4.

Analogous to Fig. 3.6, Fig. 3.8 evaluates the effect of $U_g/U_{ti,325\mu\text{m}}$ (Fig. 3.8a) and $(U_g - U_{ti,325\mu\text{m}})$ (Fig. 3.8b) on TDH for Geldart Group B particles. In Fig. 3.8a and Fig.

3.8b, eight correlations [11, 23, 32-34, 36, 44, 48] predict invariance of TDH with respect to $U_g/U_{ti,325\mu m}$ or $(U_g - U_{ti,325\mu m})$, 14 correlations [4, 5, 22, 27, 29, 30, 35, 40, 45, 46, 49, 85] predict that TDH increases with both $U_g/U_{ti,325\mu m}$ and $(U_g - U_{ti,325\mu m})$, and one correlation [28] predicts negative TDH values hence is omitted from the graphs. Huge discrepancies of up to four orders of magnitude are observed between the minimum and maximum TDH values at the higher $U_g/U_{ti,325\mu m}$ (Fig. 3.8a) or $(U_g - U_{ti,325\mu m})$ (Fig. 3.8b), which imply better predictive capability for lower U_g values. Two correlations warrant noting here. First, similar to the continuous PSD systems (Fig. 4 and Fig. 5), Fournol et al. [5] also predicts the highest outlying TDH values for the monodisperse Geldart Group B cases (Fig. 3.8a and Fig. 3.8b), due to the inflation of entrainment caused by the trickle-valve used. Note Fournol et al. [5] gives relatively high TDH values but not the highest for Geldart Group A particles (Fig. 3.6a and Fig. 3.6b). Second, regardless of Geldart Groups (namely, A or B), extent of polydispersity (i.e., monodisperse or variations of PSD widths of continuous PSDs), and operating parameter (i.e., U_g), the semi-empirical Zenz and Weil correlation [4] consistently gives the lowest TDH prediction. Two possible root causes for this trend are the use of (i) a two-dimensional column which hinder the bubble movement, and (ii) Stokes modified drag correlation which gives a lower drag force, both of which leads to lower predictions of TDH values.

By comparing Fig. 3.8a (monodisperse Geldart Group B particles) and Fig. 3.4 (continuous PSDs of Geldart Group B particles), two interesting observations are noted. First, the trends of TDH with respect to $U_g/U_{ti,325\mu m}$ are largely similar for both monodisperse (Fig. 3.8) and continuous PSD (Fig. 3.4) systems. Second, in contrast to Geldart Group A whereby the TDH values of continuous PSD systems are generally higher than that for monodisperse systems, the TDH values of monodisperse systems (Fig. 3.8) are instead lower than the continuous PSD systems for Geldart Group B

particles (Fig. 3.4). This may stem from the acknowledged varying extents of inter-species momentum transfer in the polydisperse systems [63, 64].

Fig. 3.8. Predicted TDH values for monodisperse Geldart Group B particles (glass

beads) as a function of (a) $U_g/U_{ti,325\mu m}$ and (b) $U_g - U_{ti,325\mu m}$.

Analogous to Fig. 3.7, Fig. 3.9 shows the influence of d_{pi} on TDH at two fixed U_g/U_{ti} values for monodisperse Geldart Group B particles.

Fig. 3.9. Predicted TDH values for monodisperse Geldart Group B particles (glass beads) as a function of d_{pi} for $U_g/U_{ti} =$ (a) 1.01 and (b) 2.00.

An interesting observation on contradictory TDH trends predicted by two correlations developed at the lowest d_{pi} by the same authors with the same experimental data [28, 29] warrants mention in Fig. 3.9. On one hand, Sciazko et al. (1991) [29] predicts the *highest* TDH at the lowest d_{pi} for both U_g/U_{ti} (Fig. 3.9). On the other hand, Sciazko et al. (1988) [28] predicts the *lowest* TDH at the lowest d_{pi} value for the higher U_g/U_{ti} (Fig. 3.9b); note only one data point from the Sciazko et al. (1988) [28] correlation is shown in Fig. 3.9b at the higher U_g/U_{ti} . This is because the TDH values predicted

become negative at higher d_{pi} values in the slugging regime. The difference between the two correlations is that, the 1988 Sciazko et al. correlation [28] is semi-empirical and based on the assumption that TDH equals to the freeboard height where bubble fraction becomes 0.5, whereas the 1991 Sciazko et al. correlation [29] was obtained by modifying the semi-empirical Soroko et al. correlation [46] to fit their experimental data with wider PSDs in 1988 [28]. Although both sets of experimental data by Soroko et al. [46] and Sciazko et al. (1998) [28] are based on the measurement of the maximum splash height of particles, Soroko et al. [46] used a column diameter 10 times larger than Sciazko et al. (1988) [28]. This highlights the importance of generating more robust experimental datasets whose results can be reasonably extrapolated and a more unified physical understanding in future development of TDH correlations.

Five noteworthy results are apparent from the comparison of Fig. 3.7 (monodisperse Geldart Group A) and Fig. 3.9 (monodisperse Geldart Group B). First, all the correlations show the same trends of TDH versus d_{pi} for both monodisperse Geldart Group A and B particles. Generally, the available TDH correlations can be divided into three groups based on the relationship between TDH and d_{pi} at constant U_g/U_{ti} , namely, (i) TDH increases steeply at lower d_{pi} values but plateaus at higher d_{pi} values [11, 23, 32-34, 36, 44, 48], (ii) TDH is non-monotonic (i.e., increases then decreases) with respect to d_{pi} [4, 35, 46], and (iii) TDH decreases with d_{pi} [29, 45]. Second, the discrepancies between the minimum and maximum TDH values are lower for monodisperse Geldart Group B (Fig. 3.9; four orders of magnitude) than Geldart Group A (Fig. 3.7; five orders of magnitude). Similar to the observation made for the continuous PSD system, the predictive capability of the TDH phenomenon seems to be better for Geldart Group B particles than for Geldart Group A particles. Third, the same three correlations [30, 35, 46] show contradictory trends depending on whether ($U_g -$

U_{ti}) or U_g (plot not shown) is kept constant for both monodisperse Geldart Group A and Group B systems; specifically, they predict increasing [30] or non-monotonic trends (i.e., increases then decreases) of TDH [11, 23, 32-36, 44, 46, 48] with d_{pi} at constant ($U_g - U_{ti}$), but decreasing trend at constant U_g . Forth, different TDH trends for monodisperse Geldart Group A and B particles were predicted by 10 correlations [11, 23, 28, 32-36, 44, 48] at constant ($U_g - U_{ti}$). For example, eight correlations [11, 23, 32-34, 36, 44, 48] predict that TDH increases with d_{pi} for monodisperse Geldart Group A but is invariant for monodisperse Geldart Group B particles at constant ($U_g - U_{ti}$). Fifth, contrary to Geldart Group A whereby the largest discrepancies among correlations are observed at the lowest d_{pi} (Fig. 3.7), similar discrepancies are observed for the entire d_{pi} range considered for Geldart Group B (Fig. 3.9). This further underscores the need to adequately incorporate inter-particle cohesive effects into TDH correlations to allow for wider applicability.

3.2.3. Binary mixtures

- **Geldart Group A particles**

A binary mixture consists of two species with different d_{pi} and/or ρ_p . Although it also falls under the category of polydisperse systems, the behavior of binary mixtures is known to be different from continuous PSDs [93, 103, 104], hence warrants an improved understanding too. In these two sections, the available TDH correlations are evaluated for binary size mixtures (i.e., consist of two species of different d_{pi}) and binary density mixtures (i.e., consist of two species of different ρ_p). To allow for a fairer comparison between the two binary mixtures, the ratio of the U_{ti} values of the constituent species are kept the same [54]. For the Geldart Group A particles in this section, the

ratio of U_{ti} between the finer and coarser species for both binary size and binary density mixtures are fixed at 2.97.

Table 3.3 shows the base case parameters for binary size Geldart Group A particles, specifically composed of 40 μm and 75 μm FCC particles. Fig. 3.10 shows TDH as a function of the mass fraction of the finer species in a binary size mixture of Geldart Group A particles at two different $U_g/U_{ti,75\mu\text{m}}$. All of the correlations, except Sciazko et al. (1991) [29] and Ko [45], predict that TDH decreases with mass fraction of the finer species, in part due to the more pronounced inter-particle cohesive interactions for smaller Geldart Group A particles. Note that elutriation flux is known to increase with the composition of the fine species (which belong to Geldart Group A or C) above a critical size, or else the elutriation flux has been observed to plateau or even decrease due to the dominance of inter-particle cohesion [102, 105, 106]. The relationship between TDH and the elutriation flux value is not so straightforward, since the former is based on the rate of change of elutriation flux with height; however, the common thread lies in that predictive capability of both is hampered by the inability to quantify the effect of inter-particle cohesion [54]. Sciazko et al. (1991) [29] and Ko [45] predict an opposite trend to the other correlations due to the *ad hoc* inclusion of the U_g/d_{pi}^2 term to possibly account for the cohesive effects of the Geldart Group A systems investigated, but the inclusion of non-physically-based terms as such limits the utility of the correlations.

Fig. 3.10. Predicted TDH values for binary size Geldart Group A particles (FCC) as a function of mass fraction of finer particles for $U_g/U_{ti,75\mu m} =$ (a) 1.01 and (b) 2.00.

The huge discrepancies between the minimum and maximum TDH values in Fig. 3.10 of up to five orders of magnitude are due largely to three outlying high TDH predictions [29, 46] and two outlying low TDH values [4, 49]. These five outlying correlations [4, 29, 46, 49] have been similarly highlighted for continuous PSD and monodisperse systems of both Geldart Groups A and B. The common trait among these correlations is the distinctly different experimental setups and assumptions used to develop the TDH correlations compared to other correlations. For the outlying high predictions, Soroko et al. [46] employed a fluidized bed column of at least five times

that of the fluidized bed column diameter used by the other researchers, while Sciazko et al. (1991) [29] was obtained from the modification of the Soroko et al. correlation [46] based on new experimental data [28]. For the outlying low predictions, while Zenz and Weil [4] used a two-dimensional fluidized bed column and uniquely used the modified Stokes correlation, Brems et al. [49] uniquely assumed that the initial particle velocity is equal to the bubble velocity at the bed surface. Collectively, the five outlying correlations indicates the lack of a unifying platform for both the experimental and modeling aspects of TDH is the root cause of the orders-of-magnitude discrepancies.

Table 3.3 shows base case parameters for binary density Geldart Group A mixtures, consisting of FCC particles as the finer species ($\rho_p = 1450 \text{ kg/m}^3$) and glass beads as the coarser species ($\rho_p = 2500 \text{ kg/m}^3$).

Along the same vein as Fig. 3.10, Fig. 3.11 displays TDH as a function of mass fraction of the finer species (FCC) at two different $U_g/U_{ti, \text{glass beads}}$. With the increase of mass fraction of FCC, all the correlations predict decreasing TDH values, except Sciazko et al. (1991) [29], which predicts that TDH increases with mass fraction of FCC due again to the U_g/d_{pi}^2 term. Similar to Fig. 3.10, discrepancies of up to five orders of magnitude exist between the minimum and maximum TDH values, and the same five correlations persist to give outlying TDH predictions [4, 29, 46, 49].

Fig. 3.11. Predicted TDH values for binary density Geldart Group A particles (FCC and glass beads) as a function of mass fraction of FCC for $U_g/U_{ti,glass\ beads} =$ (a) 1.01 and (b) 2.00.

• Geldart Group B particles

Similar to Table 3.3, Table 3.4 lists the base case parameters for binary size Geldart Group B mixtures, consisting of glass beads of different d_{pi} (namely, 225 μm and 500 μm). For both binary size and binary density Geldart Group B mixtures, the ratio of the U_{ti} values of the constituent species is kept at 2.2.

Fig. 3.12 presents TDH as a function of mass fraction of the finer species for the binary size Geldart Group B mixture at two $U_g/U_{ti,500\mu\text{m}}$.

Fig. 3.12. Predicted TDH values for binary size Geldart Group B particles (glass beads) as a function of mass fraction of finer particle for $U_g/U_{ti,500\mu m} =$ (a) 1.01 and (b) 2.00.

In Fig. 3.12, Fournol et al. [5] and Zenz and Weil [4] continue to give outlying high and low TDH predictions, respectively, as is the case for all the Geldart Group B base parameters tested (namely, continuous PSD, monodisperse, binary size) so far. Detailed analysis of Fig. 3.12 shows that, as the mass fraction of the finer species increases, eight correlations [11, 23, 32-34, 36, 44, 48] predict that TDH is constant, five correlations [29, 35, 45, 46] predict that TDH increases, and 9 correlations [4, 5, 22, 27, 30, 40, 47, 49, 85] predict that TDH decreases. Comparison of Fig. 3.12 (Geldart

Group B, binary size) and Fig. 3.10 (Geldart Group A, binary size) reveals two differences between Geldart Groups A and B, namely (i) less discrepancies exist among TDH predictions for Geldart Group B compared to Geldart Group A, specifically four orders of magnitude for Geldart Group B and five orders of magnitude for Geldart Group A, and (ii) the largest discrepancies among TDH predictions are observed at the *lower* mass fraction of the finer species for Geldart Group B but at the *higher* mass fraction of the finer species for Geldart Group A. This observation is consistent with that for entrainment [54].

For the Geldart Group B binary density mixture, the finer species is polystyrene (PS) with $d_{pi} = 500 \mu\text{m}$ and $\rho_p = 1050 \text{ kg/m}^3$, while the coarser species is glass beads with $d_{pi} = 500 \mu\text{m}$ and $\rho_p = 2500 \text{ kg/m}^3$. Base case parameters are displayed in Table 3.4.

The TDH values predicted by the various correlations for the binary density Geldart Group B mixtures are depicted in Fig. 3.13. Similar to Fig. 3.12 (Geldart Group B, binary size), Fig. 3.13 illustrates that the same eight correlations [11, 23, 32-34, 36, 44, 48] (which are based on bubble parameters) predict that TDH is invariant with respect to the mass fraction of PS. Compared to binary size, the binary density system has fewer correlations [29] predicting that TDH increases but more correlations [4, 5, 22, 27, 30, 35, 40, 45-47, 49, 85] predicting that TDH decreases as the mass fraction of PS increases. Binary size and binary density mixtures have been observed to behave differently [88, 93, 107], although a clear physical understanding and associated quantification remains elusive. Consistent with the observations from the comparison of binary size mixtures for the two Geldart Groups (Fig. 3.10 and Fig. 3.12), for the binary density mixtures, lesser discrepancies among TDH predictions exist for Geldart Group B (Fig. 3.13) relative to Geldart Group A (Fig. 3.11) and the larger discrepancies appear at opposite ends of the mass fraction of finer species for the two Geldart Groups.

Fig. 3.13. Predicted TDH values for binary density Geldart Group B particles (PS and glass beads) as a function of mass fraction of PS for $U_g/U_{ti, glass\ beads} =$ (a) 1.01 and (b) 2.00.

3.2.4. Effect of column diameter

• Geldart Group A particles

In the earlier sections, the evaluation of the impact of several important parameters in fluidized beds, such as Geldart Groups, range of particle sizes and types (i.e., continuous PSDs, monodisperse, and binary mixtures), and the key control parameter (i.e., U_g) on TDH value has been carried out. To extend the review to reactor configuration, this and the following sections investigate the impact of fluidizing

column diameter (D_c) on the predictive capability of TDH correlations. The reason for a closer inspection of D_c is because several authors have affirmed that effect of column diameter on TDH is significant [4, 5, 16, 22, 108], including Frantz and Juhl [22], who reported that D_c is one of three most important factors affecting TDH along with U_g and d_{pi} .

The influence of D_c on TDH hinges on the significance of the wall effect [16], which is more pronounced for smaller D_c . Specifically, a smaller D_c increases the diffusion of fine particles from the upward flow in the center of the column to the downward flow in the falling film near the walls [11, 109, 110], hence reduces TDH. On the other hand, a larger D_c allows for larger bubbles with higher bubble velocities, hence increases TDH [23]. Notably, a critical D_c exists beyond which the fluid flow transitions from laminar to turbulent and the wall effect become negligible [5, 21, 108].

In this section, TDH predictions for a range of D_c values are studied. Details of the base case parameters used in this section are listed in Table 3.3. Fig. 3.14 presents the TDH predictions for monodisperse Geldart Group A particles (namely, FCC with $d_{pi} = 80 \mu\text{m}$ and $\rho_p = 1450 \text{ kg/m}^3$) for D_c in the range of 0.1 - 2 m at two $U_g/U_{ti,80\mu\text{m}}$ (i.e., 1.01 and 2.00). As expected from previous sections, huge discrepancies of up to four orders of magnitude exist among the TDH predictions. Eight correlations [5, 27, 29, 35, 40, 45, 46], which are based only on U_g , predict that TDH is independent of D_c ; furthermore, 10 correlations [4, 11, 28, 30, 32-34, 36, 44, 48] predict independent of D_c only when $D_c > 0.6 \text{ m}$, which agrees with available experimental data [56, 111, 112]. Fifteen correlations [4, 11, 22, 23, 28, 30, 32-34, 36, 44, 47-49, 85] predict that TDH increases with D_c , with some predicting one [4, 11, 22, 23, 30, 32-34, 36, 44, 47, 48, 85] to two [28] orders of magnitude increase in TDH. Zenz and Weil [4] is the only correlation whereby TDH decreases with D_c , presumably due to the use of a two-

dimensional column as indicated earlier. In a subsequent publication by the same author, Zenz and Othmer [111] predicts that TDH increases with D_c . The inadequacy of the available TDH correlations to account for D_c highlights the importance of taking reactor configuration in the development of more physically-based TDH correlations.

Fig. 3.14. Predicted TDH values for Geldart Group A particles (FCC) as a function of

D_c for $U_g/U_{ti,80\mu m} =$ (a) 1.01 and (b) 2.00.

• Geldart Group B particles

Analogous to the previous section, this section evaluates the effect of D_c on TDH predictions for monodisperse Geldart Group B particles. Details of the base case parameters used in this section can be found in Table 3.4.

Fig. 3.15 shows the TDH predicted by existing TDH correlations for monodisperse Geldart Group B system as described in Table 3.4 for D_c of 0.1 – 2 m, both at $U_g/U_{ti,325\mu m}$ of 1.01 and 2.00. Similarly, discrepancies of up to five orders of magnitude exist among the TDH correlations. The same eight correlations [5, 27, 29, 35, 40, 45, 46] predict that TDH is invariant with respect to D_c in both Fig. 3.14 and Fig. 3.15, because the only variable governing TDH in these correlations is U_g . Furthermore, of the 12 correlations [4, 11, 23, 30, 32-34, 36, 44, 47-49] which predict that TDH increases then plateaus with D_c , four correlations [4, 11, 44, 48] present a steep increase of TDH at $D_c = 1$ m due to similar parameters in the correlations (namely, $[(2U_{bi})^2/(2g)]$) demarcating the transition from the slugging to bubbling regimes. Interestingly, all 12 of these correlations [4, 11, 23, 30, 32-34, 36, 44, 47-49] predict that TDH become constant at $D_c \geq 1.3$ m, whereas 10 correlations [4, 11, 28, 30, 32-34, 36, 44, 48] predict that TDH plateaus instead at $D_c \geq 0.6$ m for Geldart Group A (Fig. 3.14); presumably, the larger Geldart Group B particles with the corresponding larger bubble sizes result in a larger column requirement to eliminate the wall effect. On the contrary, two correlations [22, 85] predict that TDH decreases then plateaus with D_c (Fig. 3.15), although these same two correlations predict instead that TDH increases then plateaus with D_c for monodisperse Group A particles (Fig. 3.14).

Fig. 3.15. Predicted TDH values for Geldart Group B particles (glass beads) as a function of D_c for $U_g/U_{ti,325\mu m} =$ (a) 1.01 and (b) 2.00.

3.3. Conclusions

A review of the 25 TDH correlations available in literature has been conducted. Specifically, the focus is on illustrating the capability of existing correlations to predict the impact of Geldart Group classification, monodispersity or polydispersity (i.e., continuous PSDs, monodisperse particles, and binary mixtures), the key operating parameter (i.e., U_g), and reactor diameter (D_c).

Four observations are worth noting. First, among the TDH values predicted by the various correlations, discrepancies of up to five orders of magnitude and

contradictory trends are found, even if the correlations are applied only to the same Geldart Group they were developed for. The disagreement among the correlations is primarily due to the use of empirical constants and the *ad hoc* inclusions of certain terms to data-fit experimental results, which in turn is rooted in the lack of a more physical understanding of the TDH phenomenon. Note both semi-empirical and theoretical models [49] available are not amenable to wider utility due to the underlying data-fitting and assumptions related to the gap in the knowledge base. For example, the simple ballistic model [4, 23, 34, 36, 49] unsoundly neglects inter-particle interactions, while the Brems et al. [49] theoretical model is based on an assumption of initial particle velocity, the value of which has been refuted experimentally [14, 23, 43, 44, 58-60]. Second, unphysical TDH trends are predicted, namely (i) negative values of TDH for $U_{ti} < U_g$ [43], (ii) decrease of TDH with respect to U_g [28, 47], (iii) invariance of TDH as U_g increases, particularly in the slugging regime [11, 23, 32-34, 36, 44, 48], and (iv) negative or undefined TDH values for the slugging regime [28]. Further investigation reveals the reasons underlying such unphysical trends lie in (a) the use of two-dimensional columns [4, 43], which is not representative of the TDH phenomenon in three-dimensional columns due to the dominant wall effects particularly on bubble behaviors, (b) the use of bubble characteristics (d_{bi} or U_{bi}) [11, 23, 32-34, 36, 44, 48] or U_g [5, 27, 29, 35, 40, 45, 46] without accounting for other fluidized bed characteristics in the development of TDH correlations, and (c) the negative sign [28, 43] or negative power [47] of U_g , which leads to negative TDH values when the system is slugging. Third, better agreement among the correlations exists when correlations developed for Geldart Group B particles are applied only to Geldart Group B particles, compared to that for Geldart Group A, presumably due to the deficient understanding and miss-accounting of inter-particle cohesive effects more pronounced for the latter. The

dominant influences in Geldart Group A systems are likely different from those in Geldart Group B, as evident in: (i) largest discrepancies among the correlations at the lowest d_{pi} for the monodisperse Geldart Group A test case, but similar discrepancies among the correlations through the d_{pi} range considered for the monodisperse Geldart Group B test case, (ii) the predicted TDH values are higher for the continuous PSD relative to monodisperse for Geldart Group A, but vice versa for Geldart Group B, and (iii) larger discrepancies among the correlations at the *higher* mass fraction of the finer species in binary mixtures of Geldart Group A, but at the *lower* mass fraction of the finer species in binary mixtures of Geldart Group B. Forth, the study of the TDH phenomenon is lacking for the slugging regime relative to the freely-bubbling regime.

Both this review and an earlier review on entrainment [54] underscore the same need for more mechanistic-based correlations. To reduce the dependency on empirical constants and *ad hoc* parameters, which restrict wider applicability, three gaps in the current knowledgebase have to be addressed: (i) inter-particle interactions, namely, inter-species momentum transfer and inter-particle cohesive forces, (ii) accounting for clustering hydrodynamics if present, and (iii) a comprehensive, reliable experimental data set containing TDH results of various PSD widths, fluidization regimes (i.e., freely-bubbling and slugging regimes), and reactor configurations.

Nomenclature

- A = cross-sectional area of fluidized bed column, m^2
 A_0 = cross-sectional area of distributor, m^2
 $Ar_i = \frac{gd_{pi}^3\rho_g(\rho_p-\rho_g)}{\mu_g^2}$ = Archimedes number of size d_{pi} particle, -
 a, b = variables in Brems et al. [49], -
 a_c = exponential decay constant, -
 a_{cf} = cluster flux exponential decay constant, -
 C_d = drag coefficient, -
 D_c = inner diameter of fluidizing column, m

d_{b0} = initial bubble size at distributor, m
 d_{bi} = bubble size diameter, m
 d_{bm} = maximum bubble size, m
 d_{bs} = bubble diameter at bed surface, m
 d_{pi} = mean particle size of species i , m
 $Fr_i = \frac{U_g^2}{gd_{pi}}$ = particle Froude number of size d_{pi} particle evaluated at U_g , -
 g = gravitational acceleration, m/s^2
 H_c = height of fluidized bed column, m
 H_d = height from the distributor, m
 H_s = height of settled fluidized bed, m
 K_0 = entrainment rate from the bed surface, $kg/m/s^2$
 K_∞ = entrainment rate above TDH, $kg/m/s^2$
 $Re_{pi} = \frac{U_g d_{pi} \rho_g}{\mu_g}$ = particle Reynolds number of size d_{pi} particle evaluated at U_g , -
 U_{bi} = bubble velocity, m/s
 U_{ci} = cluster ejection velocity, m/s
 U_g = superficial gas velocity, m/s
 U_{mf} = minimum fluidization velocity, m/s
 U_{mi} = particle balancing velocity, m/s
 U_{psi} = particle velocity at bed surface, m/s
 U_{ri} = slip velocity, m/s
 U_{ti} = terminal velocity of species i , m/s
 u'_{vo} = constant of Horio et al. (1980) [34], -
 V = constant of Do et al. [43], -

Symbols and Greek Letters

α = constant of Wen and Chen [16], -
 σ = standard deviation of Gaussian PSD, -
 β = turbulent decay constant, -
 ε_{mf} = voidage at minimum fluidization condition, -
 μ_g = gas viscosity, $kg/m/s$
 η = constant of Horio et al. [34], -
 ρ_g = gas density, kg/m^3
 ρ_p = particle density, kg/m^3
 ϕ = particle sphericity, -

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CHAPTER 4: CLUSTER CHARACTERISTICS OF MONODISPERSE AND POLYDISPERSE SYSTEMS IN CIRCULATING FLUIDIZED BED (CFB) RISERS

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Abstract

Particle clusters are well acknowledged to affect the hydrodynamics and overall performance of gas-solid fluidized beds. Since one of the first reports on the clustering phenomenon in 1948, the understanding of particle clustering has been rigorously attempted via both modeling and experimental efforts, with significant traction gained especially in the last few decades. Accordingly, the current review targets at providing a comprehensive landscape of the experimental cluster trends to summarize the findings, particularly in circulating fluidized bed (CFB) riser to date. More questions than answers seem to have sprouted from the abundant experimental data available, which impedes model development. The quantitative comparison of cluster characteristics across studies must be treated with caution, because of (i) the impact on cluster characteristics by an interplay of a host of factors, hence the influence of a single parameter is not straightforward, even within the same study; (ii) different riser configurations, instruments, and analysis methods, which can lead to discrepancies of an order-of-

magnitude; (iii) the irregularity in the form of clusters, hence the definition and/or measurement of the various cluster characteristics differ; and (iv) the general lack in the reporting of the actual particle size distribution.

Keywords: cluster characteristics; circulating fluidized bed riser; polydispersity; particle properties

4.1. Introduction

Particles exist as both dispersed individual particles and clusters or agglomerates (i.e., groups of particles) in gas-solid fluidization systems. It is important to distinguish between particle cluster and particle agglomerate, although the two terms tend to be used interchangeably. While clusters are due to hydrodynamics effects and weak inter-particle forces, agglomerates are bonded or caked arrays of particles [1]. The focus of the current study is on particle clusters. Up to 75 % of the particles in the freeboard exist as clusters in some systems [2], which underscores the importance of understanding this phenomenon as this inevitably impacts the overall performance of gas-solid fluidized beds. Because particle clusters exhibit larger characteristic particle sizes and smaller local void fractions compared to dispersed particles [3-6], the efficiency of gas–solid contacting, which underlies the widespread use of fluidized bed in industrial processes, is affected. Parameters affected span particle slip velocity, drag force [7], pressure drop [7, 8], local voidage [4], mixing [9], and heat and mass transfer [10, 11].

The particle clustering phenomenon was first reported in 1948 [12], and attempts at understanding discrepancies between expected slip velocities determined from the terminal velocity and what was measured have ensued. Since then, investigations in this area have resulted in more questions than answers. As seen in Fig. 4.1, the number of

publications on particle clustering has been steadily increasing in the past few decades. The total number of publications on clustering has inflated more than 2.5 times in the decade of 2006 – 2015 compared to 1996 – 2005. Interestingly, although experimental efforts used to surpass modeling efforts before year 2000, the rapid advances in computing capability in the 21st century has driven modeling to be approximately twice that of experimental efforts in the subsequent 5-year period of 2001 – 2005. In the most recent decade of 2006 – 2015, the number of modeling efforts is clearly more than twice that of the experimental ones. Experimental and simulation efforts are complementary components in the quest for more understanding on the clustering phenomenon; experimental data is needed for model validation, while the model in turn provides for the extraction of physical understanding. Unfortunately, the abundant experimental results available appear to give contradictory trends with respect to key parameters like particle properties and operating conditions. As indicated by previous reviews [13, 14], experimental data tend to not perform well beyond the limited scope of experimental conditions tested, which at least in part contributes to the contradictory trends. Therefore, the current study aims to summarize the experimental understanding of cluster characteristics in Circulating Fluidized Bed (CFB) risers and attempts at extracting some of the dominant underlying physics.

Fig. 4.1. Publications related to particle clustering in gas-solid fluidized beds. Source = Google Scholar; keyword search = cluster OR strand OR clustering AND particle OR Geldart OR fluidization OR fluidized OR CFB OR riser OR downer OR gas solid; retrieved = 7 April 2016.

4.2. Literature Review

4.2.1. Methods to characterize particle clusters

- **Measurement of cluster characteristics in experiment**

A host of methods, which has been used to examine particle clusters experimentally, can broadly be categorized into non-invasive versus invasive, and optical versus non-optical methods. Non-invasive techniques are superior notably for the lack of disruption to the hydrodynamics being characterized, although difficulties with data interpretation and lower level of discrimination are recognized [15-18]. On the other hand, invasive techniques inevitably disrupt the flow, but are expedient for providing localized information. Optical methods, typically based on absorption or transmittance of visible light or infrared wavelengths in the electromagnetic spectrum, can reveal useful information about particulate behavior, whereas the non-optical

methods typically rely on acoustic (e.g., accelerometer [19, 20]) or electrical (e.g., capacitance probe [21-26]) signal. Optical methods can be non-intrusive (e.g., naked eye visualization [27], microscope [28], laser sheet technique [6, 29], high-speed video camera [2, 11, 29-32], Particle Imaging Velocimetry (PIV) [33], Laser Doppler Velocimetry (LDV) [34], and Phase Doppler Particle Analyzer (PDPA) [35]) or intrusive (e.g., fiber optic probe [30, 32, 36-47]). The shortcoming of optical techniques lie in that the shallow visible light penetration depth and light diffraction (e.g., on the curve surface of the wall column, or transparent or reflective particles like glass beads) limit their applicability to low solid concentration [17].

Regarding the interpretation of the experimentally obtained signals, the signal analysis method plays an indelible role in the results. Because clusters appear as regions of higher concentrations, most data analysis methods hinge on either using the signals directly or translating the signals into indications of concentrations, then applying a threshold value above which clusters are identified. A time-invariant ‘heterogeneity index’ [48, 49] has been coined to determine cluster parameters based on the Soong et al. [25] criteria, whereby a cluster is deemed to exist when (i) the solid concentration is greater than the time-averaged local solid concentration, and (ii) the cluster size (i.e., based on the duration in which the solid concentration is greater than the time-averaged solid concentration) is at least a hundred times that of the particle diameter (d_p). The threshold criteria (i) was subsequently modified several ways such that clusters are presumed to exist only when the solid concentration is greater than the time-averaged local solid concentration *plus* multiples (n) of the standard deviation of the signal (σ), in which n has been designated one [21], two [26, 32], three [25], or other factors depending on superficial gas velocity, solid mass flux, and axial or radial position [35, 36]. Furthermore, because it was observed that the experimentally obtained cluster data

was not normally distributed [38], which thereby indicated that the mean and standard deviation are meaningless, various percentiles (e.g., 50th percentile [50]) were instead proposed as more accurate thresholds. Another method of identifying threshold values is wavelet decomposition [51], which provides a non-linear, multi-scale means of characterizing clusters [52]. Clusters were assumed to be found at and above the 50th percentile [38, 39] of the reconstructed voltage signal, wherein the high-frequency noise and low-frequency equipment-scale effects were disregarded. Subsequently, Yang and Leu [47] further employed the wavelet analysis technique to determine an appropriate level of approximation sub-signal as a threshold for cluster identification, which allowed for time-dependent threshold values. This method was employed to characterize clusters in both dense and dilute risers, and polydisperse particle systems [44, 45, 53].

- **Prediction of cluster characteristics from computational study**

Detailed gas and particle behaviors (e.g., inter-particle forces, solid dispersion) are difficult, if not almost impossible, to obtain from laboratory experiments. Simulation efforts can bridge this gap. Both the two types of numerical simulations, namely, the Two-Fluid Model (TFM) and the Computational Fluid Dynamics-Discrete Element Method (CFD-DEM), have been used for studying clusters. In the TFM, or Eulerian-Eulerian approach, both the gas and particle phases are treated as continuums. Particulate flows are described by the modified Navier-Stokes equations, which account for inelastic inter-particle interactions and require additional closure models for solid pressure and viscosity (e.g., Kinetic Theory Granular Flow (KTGF) [54]). Such closure models can be developed empirically from experimental data or theoretically-verified from Direct Particle Modeling (DPM) simulation of granular flow. Numerical analysis on cluster in CFB can be traced back to 1989, when Sinclair and Jackson [55] found via a RANS (Reynolds Averaged Navier-Stokes) model higher particle concentration near

wall, which corresponded to clusters. Subsequently, Tsuo and Gidaspow [56] carried out a TFM investigation in a CFB riser and found that clusters form due to gas velocity gradients. Subsequently, TFM simulations using cluster diameters as the characteristic particle diameter showed better agreement with experimental results [4]. TFM simulations require less computational time and capacity compared to CFD-DEM simulations, which is an Eulerian-Lagrangian approach; however, the choice of appropriate closures remains more an art than a science [57]. The CFD-DEM approach uses continuum gas flow and discrete particle flow. Individual particle movement is evaluated at the particle-grid level using DPM simulations, e.g., Monte Carlo [58, 59], cellular automata [60], or Direct Element Method (DEM) [61]. There are two models to evaluate inter-particle interactions, namely, hard-sphere (i.e., binary and instantaneous particle collisions) [62] and soft-sphere (i.e., multiple particle collisions) [63] approaches. While the hard-sphere model can more precisely resolve particle contact, the soft-sphere model provides enhanced computational speed by sacrificing precision [63].

The choice of appropriate models and assumptions determine the accuracy of simulation results [64]. Tsuji et al. [58] found that the cluster population is larger in CFD-DPM compared to TFM, despite similar trends with respect to the impact of superficial gas velocity, solid flux, and particle size. Particle shape [65, 66], cluster shape (e.g., spherical [4, 67, 68] or cylindrical [9]), column dimensions (e.g., two-dimensional column [4, 9, 65, 66, 69-71]), the type of forces considered (e.g., inter-particle [72, 73], slip/shear or Saffman [9, 69, 71], and slip/rotation or Magnus [69, 71] forces) cause differences among simulation results, and between simulation and experimental results. Other factors more related to the operating conditions also lead to discrepancies among simulation results, including solid velocity and concentration

distributions [65, 74-83], solid segregation and dispersion [84, 85], flow pattern and geometrical configuration [80, 82, 86-88], particle rotation [89], solid circulation [90, 91], residence time distribution [92], and gas-solid back-mixing [93-95], among others. Multiple challenges persist for numerical investigations of particle clustering, such that the simulation of clusters in a more practical three-dimensional full-loop CFB riser remains a goal to be achieved. The inclusion of clustering behaviors will improve the increasing number of reports on full-loop modelling of gas-solid motion in CFBs via both TFM [65, 79, 81, 83, 90, 95-97] and CFD/LES-DEM (whereby LES stands for Large Eddy Simulation) coupling approaches [98].

4.2.2. The origin of particle clusters

The origin of particle clusters lies in the interaction of the particles with their surroundings, either with other particles or the fluid or even in a vacuum. Notably, clustering of glass beads with a particle diameter of 100 μm has been reported to exhibit clustering even in vacuum (i.e., in the absence of hydrodynamic forces) [99]. It is important to understand the mechanism of cluster formation and the corresponding driving force that affects the characteristics of clusters. Particle clusters have been observed near bubbles [2] due to the gas velocity fluctuations (up to 1 m/s with frequency of 20 Hz [9]) and inter-particle collisions [100, 101] induced. Inter-particle and gas-particle interactions, rather than wall friction, have been reported to play a more dominant role in particle clustering [65].

The formation of clusters is generally acknowledged to be as follows: (1) inelastic particle collisions dissipate kinetic energy, thereby decrease particle velocity (i.e., collisional cooling) [4, 102] and local gas pressure (i.e., viscous dissipation) [101, 103]; (2) the surrounding gas and particles flows toward these lower pressure regions

created to form particle clusters [99, 104, 105]. A cluster, which is denser than the dispersed particles, increases inter-particle forces and collisions, which decrease granular temperature further [2, 65, 106], thereby leading to more clustering. Conversely, the more forceful inter-particle collisions partaken by the clusters also lead to their breakup into either smaller clusters or disperse particles. Inter-particle forces and collisions exert more dominant effect in dense systems (e.g., bubbling fluidized bed), while hydrodynamics instability also plays a role in turbulent systems (e.g., near the gas distributor or in a CFB riser) [2, 43]. The following sections focus on inter-particle and gas-particle interactions, which are complementary driving forces in cluster formation, growth, and deformation.

- **Inter-particle interactions**

Although hydrodynamic forces have been reported to be the most dominant in terms of energy dissipation (which leads to clustering), followed by inter-particle cohesive forces then inter-particle collisions [43, 107], the contributions by the pervasive particle-particle interactions cannot be ignored. Because energy dissipation due to inelastic particle collisions reduces inter-particle distance and hence increases inter-particle forces [2], the role of inter-particle interactions may not be negligible relative to hydrodynamics instability in causing clustering.

Inter-particle forces refer to short-ranged particle-level forces, e.g., van der Waals (VDW), electrostatic, and capillary forces. VDW is the most important inter-particle force in fluidized bed systems [108-110] and consists of electrodynamic forces between molecules and atoms on the particle surface (i.e., Keesom, Debye, and London dispersion forces) [111], and has been found to promote particle clustering at high solid concentration, but suppress cluster formation near wall at low solid concentration [65]. VDW exists as attractive and repulsive forces, and depends on dielectric particle surface

properties [99, 112]. The net VDW force between two identical particles is always attractive in a closed system, with its magnitude depending on particle distance and net potential energy between the particles [111]. Electrostatic force typically occurs due to inter-particle or particle-wall column friction, which promotes electron transfer between the surfaces, thereby leading to interactions between the charged surfaces. Regarding particle charges, comparison of the clustering of conductive and non-conductive particles indicate that electrostatic forces did not affect clustering [99]. Capillary force (i.e., liquid bridging) has been found to promote particle clustering in cases of high relative humidity (i.e., greater than 45 %) [111].

Cluster formation has been found to be driven by nanonewton cohesive forces originating in a combination of VDW interactions and capillary bridges between nanometer-scale surface asperities [99]. Inter-particle cohesive forces are known to underlie the higher tendency of smaller particles (e.g., Geldart Group A) to form clusters [111, 113], because the higher ratio of external surface area per volume of smaller particles increases inter-particle cohesive forces relative to gravitational and drag forces [114, 115]. On the other hand, repulsive inter-particle interactions have been found via CFD-DEM simulation to decrease the clustering tendency [65].

Two types of inter-particle collisions based on the differences in the conservation of kinetic energy are well-acknowledged. Whereas inelastic collisions dissipate particle kinetic energy into frictional and viscous losses, elastic collisions preserve the total kinetic energy of the system before and after each collision [116]. Particle clustering is reported as being promoted by inelastic, rather than elastic collisions [117, 118], regardless of whether the CFB riser is dense [103] or dilute [69, 71, 119]. The extent of collisions is typically quantified as granular temperature, which has been reported to

decrease then plateau with the increase of particle collisions [2, 54, 65] and radially from the riser core to the wall [102].

Inter-particle collisions is evaluated by two parameters, namely, coefficient of restitution (typically symbolized as e and β , for normal and tangential components of contact forces, respectively) and coefficient of Coulomb friction (typically symbolized as μ for particle sliding) [120]. Regarding the coefficient of restitution, the magnitude is between 0 and 1, whereby 0 and 1 indicate completely inelastic and elastic collisions, respectively; the lower the coefficient of restitution is, the higher the collisional cooling and the tendency of particle clustering, and the denser and smaller the clusters tend to be [121]. The coefficient of restitution is both a function of the colliding material (i.e., particle, wall) properties and their relative impact velocities [122]. Although a higher coefficient of Coulomb friction reduces the probability of particle slip [71, 121], the rotating particles only form clusters if either collisional cooling or cohesive forces is insufficient [99]. Particle rotation has been reported to be primarily caused by particle collisions rather than gas shear flow [89].

Inter-particle collisions are affected by operating conditions and particle properties [2, 30, 43, 99, 123, 124]. With respect to operating conditions, higher solid loading (m), higher superficial gas velocity (U_g), and wider particle size distribution (PSD) all contribute to increasing inter-particle collisions [43, 65, 125]. As for particle properties, the influence of fine particles (i.e., particle diameter (d_p) $< 44\ \mu\text{m}$) on cluster formation has been shown via high-speed camera, whereby clustering was generally absent for fine particles although some clusters may form in the annulus [2, 43]. Furthermore, fine particles tend to be driven to the core of a riser whereas large particles are driven to the wall [126]. Thus, clustering at the wall may not be driven by fines in a riser annular region the same way it is driven in a fluidized bed [2].

- **Gas-particle interactions**

Gas-particle interactions can be described by four types of lift forces induced on the particle [127], namely, (i) Saffman force, which originates from a velocity or pressure or temperature gradient; (ii) Magnus force, which originates from particle rotation and moves the particle toward the higher-velocity region; (iii) drag force, which arises from differential velocities between the fluid and the particle, and (iv) Basset force, which arises from the acceleration or deceleration of a particle in a fluid. The Basset force is reported on one hand to be usually neglected due to large differences between gas and particle densities in gas-solid fluidization [127], but on the other hand shown to be non-negligible since experimental data suggested that clusters are formed in the acceleration region where Basset forces are significant [44, 45, 53]. Shear stress has been reported as being necessary for cluster formation either in the absence [72, 73] or presence [128] of inter-particle forces.

The impact of hydrodynamics instability effects (e.g., fluctuation of local gas velocity, gas pressure, and gas temperature) on clusters depends on cluster characteristics and operating conditions [104]. Regarding cluster characteristics, larger clusters experience higher drag forces, which shred cluster layers thereby reducing the cluster size [69], while dilute clusters (with voidage beyond 90 %) tend to be destroyed due to the increasing gas flow within the cluster [9]. As for operating conditions, the denser the cluster (i.e., higher solid concentration) is, the more inadequate hydrodynamic forces are in either growing or destroying clusters [43] due to the dampening effect of gas turbulence by particle inertia [71].

4.3. Results and Discussion

The impact of polydispersity, particle properties, and operating conditions on the lateral and axial trends of the various cluster characteristics (namely, size, duration, frequency, and appearance probability) from experimental data reported to date are summarized here. In view of the abundant experimental data on cluster characteristics in CFB risers available in literature, easing the readability of the graphs in this review necessitates two criteria for the data presented here. Specifically, within the same study, cluster characteristics were measured (i) at various axial and lateral positions within the same study, and/or; (ii) over a broad range of polydispersity, particle properties, and operating conditions. The details of the experimental parameters from previous studies used in the graphs of the current review are listed in Table 4.1. Experimental results with unspecified particle size distribution (PSD) in Table 4.1 were assumed as narrow Gaussian distributions.

Table 4.1. Experimental studies on cluster characteristics in CFB riser.

Year	Author(s)	D (m)	H (m)	Polydis persity	Geldart Group	Materi al	d_p (μm)	ρ_p (kg/m^3)	U_g (m/s)	G_s ($\text{kg/m}^2/\text{s}$)	$m = G_s$ $/(\rho_g U_g)$ (-)	Analysis Technique
1995	Li et al. [41]	0.09	10	n/a	A	FCC	54	929.5	1.31 – 3.49	5.4 – 73.64	3.5 – 17.9	Fiber optic probe (~1 s sampling time, 10 kHz), n = unspecified
1995	Wei et al. [129]	0.186	8	n/a	A	FCC	54	1398	2.27 – 3.85	58 – 164	19.2 – 37.5	1D fiber optic image analysis ((60 x 60) pixels, 500 fps, probe = 100 x 2 mm)
2000	Sharma et al. [21]	0.15	10.8	n/a	A, B	Glass beads	70 – 120	n/a	4 – 6.6	75	9.7 – 15.9	Capacitance probe (n/a), $n = 2\sigma$

2002	Manyele et al. [36]	0.076	10	n/a	A	FCC	67	1500	5.5 – 10	100 – 550	10.6 – 58.4	Fiber optic probe (60 s sampling time, 0.97 kHz), n $= 1 - 1.4\sigma$
2005	Liu et al. [130]	$L \times W$ $= 0.1$ $\times 0.1$	2.4	n/a	A	Glass beads	53	2500	0.7 – 0.85	0.75 – 1	0.9 – 1.2	Phase Doppler Particle Analyzer (PDPA), $n = 1.5 -$ 2.4σ
2007	Guenther and Breault [38]	0.3	15.3	n/a	A	Cork	812	189	3.81 – 5.2	3.57 – 17.85	0.6 – 3.3	Fiber optic probe (65.5 s, 12.5 kHz), wavelet analysis
2009	Yang and Leu [47]	0.108	5.75	n/a	A	FCC	78	1880	0.91 – 1.86	0 – 19.5	0 – 8.9	Fiber optic probe (n/a), Multi

												Resolution Analysis (MRA) with wavelet analysis
2011	Chew et al. [53]	0.184	4	Mono-, binary, continuous PSD	B	Sands, glass beads, polystyrene	165 – 327.5	1050 – 2650	1.5 – 1.7	0.05 – 0.58	0.1 – 0.2	Fiber optic probe (30 s sampling time, 1 kHz), wavelet analysis
2011	Xu and Zhu [30]	$L \times W$ = 0.114 $\times$ 0.019	7.6	n/a	A, B	FCC, glass beads, sands	67 – 288	1877 – 2498	5.5 – 8	100 – 200	15.4 – 30.9	(1) Fiber optic probe (1.31 s, 100 kHz), $n = 2\sigma$; (2) high speed video camera

												((1280 x 512) pixels, 1000 fps, image lens = 12.5 – 25 mm)
2012	Chew et al. [44]	0.3	18.3	Monodi spere	B	HDPE, glass beads	170 – 650	900 – 2500	13.5 – 17	120 – 260	5.9 – 16	Fiber optic probe (30 s sampling time, 0.1 – 1 kHz), wavelet analysis
2012	Chew et al. [45]	0.3	18.3	Binary, continu ous PSD	B	HDPE, glass beads	170 – 650	900 – 2500	10 – 17	50 – 300	2.8 – 25	Fiber optic probe (30 s sampling time, 0.1 kHz), wavelet analysis

2012	Xu and Zhu [32]	$L \times W$ = 0.114 $\times$ 0.019	7.6	n/a	A, B	FCC, glass beads, sands	67 – 288	1877 – 2498	5.5 – 8	100 – 200	15.4 – 30.9	(1) Fiber optic probe (1.31 s, 100 kHz), $n = 2\sigma$; (2) high speed video camera ((1280 x 512) pixels, 1000 fps, image lens = 12.5 – 25 mm)
2013	Kiani et al. [131]	0.05	1	n/a	B	Glass beads	150 – 300	2640	3.5 – 4.4	62 – 72	12 – 17.5	Fiber optic probe (15 s, 300 kHz), wavelet analysis

n/a = not available

4.3.1. Cluster size

Simulations require better predictions of cluster sizes to improve the calculation of drag force, pressure drop, voidage, and local gas and solid velocities [4, 132], and thereby improve the physical understanding and predictive capability of riser hydrodynamics. Unfortunately, experimental efforts targeted at revealing cluster sizes have given contradictory trends, which hinder modeling efforts and hence physical understanding of the clustering phenomenon.

To better summarize the experimental trends, Fig. 4.2 compiles cluster size data of monodisperse Geldart Groups A and B particles from the studies listed in Table 4.1. The range of solid loading ($m = G_s/(\rho_g \cdot U_g)$) evaluated ranged from 0.7 to 37.5. Other than Kiani et al. [131], which was on Geldart Group B, all the studies were on Geldart Group A. Each sub-plot in Fig. 4.2 displays cluster size as functions of radial (r/R , whereby r is the radial position evaluated and R is the entire radius of the riser, and $r/R = 0$ and 1 refer respectively to riser center and wall) or lateral (w/W , whereby w is the lateral position evaluated and W is the entire length of the riser, and $w/W = 0$ and 1 refer respectively to riser center and wall) positions depending on whether the cross-sectional area of the riser was circular or rectangular. Each of the three sub-plots in Fig. 4.2 refers to three axial positions along the riser, namely, (a) lower ($h/H \approx 0.2 - 0.3$, whereby h is the axial position evaluated and H is the entire height of the riser), (b) middle ($h/H \approx 0.5 - 0.6$), and (c) higher ($h/H \approx 0.7 - 0.8$) axial positions. It should be noted that the y-axes are in logarithmic scales.

Fig. 4.2. Lateral trends of cluster size at (a) $h/H \approx 0.2 - 0.3$, (b) $h/H \approx 0.5 - 0.6$, and (c) $h/H \approx 0.7 - 0.8$. Other than Kiani et al. [131], which was monodisperse Geldart Group B, all the studies were on monodisperse Geldart Group A particles; the ‘i’ and ‘ii’ in the legend for Kiani et al. [131] represent two particle types. Results presented here satisfy the two criteria: cluster characteristics were measured (i) at various axial and lateral positions within the same study, and/or; (ii) over a broad range of polydispersity, particle properties, and operating conditions.

• Lateral and axial trends

With regards to the radial or lateral trends, Fig. 4.2 shows that the variations in the cluster sizes reported are up to two orders-of-magnitude, with Fig. 4.2c displaying lesser discrepancies relative to Figs. 4.2a - b. Two notes on the variations are worth highlighting. Firstly, Fig. 4.2c displays only one order-of-magnitude of variation in cluster size because the data herein were obtained from a smaller range of ρ_p (i.e., 200 – 900 kg/m^3 in Fig. 4.2c versus 200 – 2500 kg/m^3 in Figs. 4.2a - b) and m (i.e., 0.6 – 18

in Fig. 4.2c versus 0.6 – 35 in Figs. 4.2a - b). However, all the sub-plots in Fig. 4.2 compare a similar range of d_p (i.e., 50 – 800 μm) in cylindrical columns (i.e., $D = 0.05$ – 0.3 m and $H = 1$ – 15.3 m), except Xu and Zhu [32] who examined two-dimensional rectangular column (with L by W of 0.114 m by 0.019 m). Therefore, it appears that the reduction in the upper limit of ρ_p and m respectively by one-third and half, and/or the possible exit effects higher in the riser were responsible for reducing the variations in Fig. 4.2c by one order-of-magnitude. Secondly, although all of measurements in Figs. 4.2a – c were obtained with optical-based equipment like a fiber optic probe or a high-speed video camera, the data acquisition frequencies and analysis techniques varied. Among experimental results obtained from fiber optic probes [32, 38, 41], sampling time varied from 1 s [32, 41] to 65.5 s [38], while sampling frequency spanned 10 kHz [41] to 300 kHz [131]. Image analysis was carried out with pictures of varying resolutions obtained with an intrusive boroscope [129] or non-intrusive high-speed video camera [32] at sampling frequencies of 500 frame per second (fps) [129] or 1000 fps [32]. It has been shown that a higher data acquisition frequency reduced the magnitude of cluster duration (and hence cluster size), although the lateral qualitative trends of cluster characteristics were unaffected [53]. Furthermore, the threshold value for identifying clusters was defined differently among available studies, such as two standard deviations [32], arbitrary values based on observations [41, 129] or frequency-based values obtained through wavelet analysis [38, 131].

Other lateral results not presented in Fig. 4.2 are described in the following. Simulations found that the maximum cluster size was near the wall at the bottom of the column [4, 103], and in the annulus at mid-height and the top of the riser [4]. This is only seen for the small clusters shown in Fig. 4.2. Other experiments [43, 133] and simulations [56, 74] agreed that radial cluster size increased for smaller column

diameters, because wall friction reduced gas turbulence and particle movement near the wall. A more uniform radial cluster size distribution was observed in a smaller column [50] or higher in the riser [44], and further found to mimic a Gaussian distribution [134].

Fig. 4.2 shows that axial position tended to have insignificant effect on cluster size regardless of solid loading (m) in the range of 0.7 – 36.2. Solid loading (m) has significantly greater impact on cluster size than axial position, as well be discussed later. Axial trends not presented in Fig. 4.2 due to different ranges of h/H are described in the following. Axially, cluster size has been found to increase [117], decrease [103, 135], or increase then decrease [43]. With regards to increasing cluster size with height, simulation results [117] indicate the underlying reason to be bubbles or jets formation at the riser bottom tending to increase gas turbulence, which led to clusters breaking up into smaller ones [103], while particles experiencing more shear stresses (i.e., higher resistance to motion due to inter-particle interaction or friction) [136] tended to accumulate with height to form larger clusters [117]. As for the opposing trend of decreasing cluster size with height, the evidence appears to indicate that the denser bottom region enhanced clustering; simulation results [4, 103, 135] indicated that bubbles and jets tended to reduce inter-particle distances especially at the denser bottom regions, hence increasing inter-particle interactions and collisional cooling, thereby promoting clustering. High-speed video camera images have proven the existence of clusters in the dense bed of Geldart Group A particles, wherein bubbles served to concentrate fine particles or existing clusters to promote particle clustering, which was further enhanced with increasing circulation rate within the bed [2]. As the clusters approached the bed surface, they attained a maximum size at the dense bed surface, then became smaller in the freeboard due to the elutriation of the clusters [43], which is consistent with findings that ascending clusters were smaller than descending ones [6,

137]. The results of Chew et al. [44, 45] using wavelet decomposition found a similar trend in that clusters formation was most dominant in the acceleration region of the riser. Regardless of whether the correlation between cluster size and height was positive or negative, the impact of axial position on the cluster sizes of monodisperse and polydisperse (i.e., binary mixtures and continuous PSD) particles was reported to be more pronounced than the impact of radial position, particle properties, and operating condition [44, 45, 53].

- **Operating conditions**

Figs. 4.2a – c show that cluster size may increase, decrease, or exhibit a non-monotonic trend with respect to r/R , depending on particle properties and operating conditions. Specifically, cluster sizes tended to decrease with r/R for dilute risers (i.e., $m < 3.5$ [38]) and increase with r/R for dense risers (i.e., $m = 20.3 – 36.2$ [129]). As for the intermediate m values, Fig. 4.2 indicates that the trends of cluster size with respect to r/R could increase, decrease, or be non-monotonic [41, 129], with the exception of the flatter profiles in the two studies wherein the riser had a rectangular cross-section with a small width (i.e., 0.019 m) in one case [32], and the narrowest diameter (i.e., 0.05 m) and also the only study on Geldart Group B in the other [131]. Evaluation of the experimental results obtained within the same studies [38, 41, 129] show no clear, consistent trends between the cluster size of the monodisperse particles and m (Fig. 4.2a – c). Furthermore, among different studies, the similar m conditions at similar axial positions gave cluster sizes varying by one order-of-magnitude (e.g., $m = 14.8$ in Li et al. [41] versus $m = 15.4$ in Xu and Zhu [32]; $m = 17.9$ in Li et al. [41] versus $m = 19.2$ in Wei et al. [129]), whereas some studies gave similar cluster sizes at similar m conditions (e.g., $m = 3.3$ in Guenther and Breault [38] and $m = 3.5$ in Li et al. [41]). While the smaller cluster sizes in Wei et al. [129], Xu and Zhu [32], and Kiani et al.

[131] appear to be related to the higher m conditions investigated, the comparison between Guenther and Breault [38] and Li et al. [41] bucks the trend in that the latter presented larger clusters for the higher m conditions. In particular, even among the consistent trends, two notes on three studies [32, 129, 131] should be flagged: (i) the smaller cross-sectional areas of the columns in Xu and Zhu (whereby width = 0.019 m) [32] and Kiani et al. (whereby diameter, $D = 0.05$ m) [131] mean that the wall effects were more significant; and (ii) an intrusive probe whose length spans more than half the column diameter was used in Wei et al. [129], which inevitably disrupted the clusters.

In other studies not presented in Fig. 4.2 due to different ranges of h/H investigated, contradictory relationships between m and cluster size were observed, i.e., cluster size could increase [34, 56, 138] or decrease [6, 139] with m . This contradiction may be related to the observation reported that an increase in m diminished micro-clusters (≤ 20 particles) and promoted macro-clusters (> 100 particles) [2, 42], hence the use of a fixed threshold across operating conditions means that such a change in the nature of the cluster would not be picked up.

Table Table 4.2 lists superficial gas velocity (U_g) and solid mass flux (G_s) values to enable the delineation of the impact of U_g , G_s , and m on the cluster size of Geldart Group A particles in Figs. 4.2a – c. For U_g in the narrow range of 3.49 – 3.81 m/s (Table 4.2) a non-monotonic correlation with respect to G_s in the range of 10.71 – 90 kg/m²/s (which corresponded to m values between 2.4 and 20.3) [38, 41, 129] was observed in that cluster size increased then decreased with G_s at $h/H \approx 0.5 - 0.6$ (Fig. 4.2b). Because the comparison spans three studies, other effects such as measurement technique or column dimensions may be at play, hence the comparison of the impact of G_s at constant U_g was

also inspected within the same study. In Li et al. [41], cluster size largely increased linearly with G_s . In Wei et al. [129], increasing G_s twofold at $U_g = 2.56$ m/s also increased cluster size up to twofold in the annulus and also near the wall. In Guenther and Breault [38], keeping U_g constant at 4.65 m/s, increasing G_s threefold and fivefold caused cluster size to increase by 2 - 3 and 2 - 5 times, respectively. Collectively, cluster size tended to increase monotonically with G_s , although the increase was lesser at a lower solid loading, m (< 3.3). However, another study in a dilute CFB riser (i.e., $m = 0.6 - 0.83$) indicated that cluster size decreased with m [6], which may be traced to the considerably lower G_s ($0.09 - 0.6$ kg/m²/s) and U_g ($0.4 - 0.6$ m/s) conditions compared to the other studies evaluated in Fig. 4.2 (see Table 4.2). Due to non-overlapping G_s values in Table 4.2, the impact of U_g at constant G_s was evaluated within the same study. For $G_s \approx 90$ kg/m²/s in Wei et al. [129], the increase of U_g from 2.95 to 3.76 m/s (Table 4.2) had a negligible impact on cluster size for $r/R < 0.4$ but led to a decrease in cluster size when $r/R > 0.4$ (Fig. 4.2b). The lack of a relationship between the cluster size of Geldart Groups A particles and U_g in the riser core was also reported previously [36]. As for the lower $G_s = 10.7$ kg/m²/s in Guenther and Breault [38], the increase of U_g from 3.81 to 5.20 m/s (Table 4.2

Table) led generally to a decrease in cluster size at all axial positions (Figs. 4.2a – c).

The inverse correlation between cluster size and U_g agrees with the results of other experiments [5, 11, 36, 140-142] and simulations [4, 65] using Geldart Group A particles. On the contrary, some other experimental results indicated contradictory trends in that an increase in U_g produced larger clusters for Geldart Groups A [137] and B [50, 137] particles. Regarding the contradictory trends, it should be noted that Mostoufi et al. [137] and Firuzian et al. [50] investigated the turbulent fluidization

regime, instead of fast fluidization or dilute transport regimes assessed in other riser studies (Table 4.2

Table).

Table 4.2. Tabulation of U_g , G_s , and m values investigated in previous studies.

Year	Author(s)	U_g (m/s)	G_s (kg/m ² /s)	m (-)
1995	Li et al. [41]	1.31	5.40	3.5
		1.31	6.01	3.9
		2.62	38.04	12.3
		2.62	42.03	13.6
		3.49	60.69	14.8
		3.49	73.64	17.9
1995	Wei et al. [129]	2.56	58	19.2
		2.56	77	25.5
		2.56	113	37.5
		2.95	89	25.6
		3.76	90	20.3
		3.85	164	36.2
2000	Sharma et al. [21]	4.0	75	15.9
		6.6	75	9.7
2007	Guenther and Breault [38]	3.81	10.71	2.4
		4.65	3.57	0.7

		4.65	10.71	2.0
		4.65	17.85	3.3
		5.20	10.71	1.7
2011	Xu and Zhu [30]	5.5	100	15.4
		5.5	200	30.9
		8.0	200	21.2
2011	Chew et al. [53]	1.5	0.24	0.1
		1.7	0.42	0.2
2012	Chew et al. [44]	13.5	120	7.4
		13.5	260	16.0
		17.0	120	5.9
		17.0	260	12.7
2012	Chew et al. [45]	10.0	50	4.2
		10.0	300	25.0
		13.5	120	7.4
		13.5	260	16.0
		15.0	50	2.8
		15.0	300	16.7
		17.0	120	5.9
		17.0	260	12.7

Because a similar solid loading (m) may give different magnitudes of local solid concentration, depending on U_g , G_s , and column configurations [36, 143], the macroscopic specification of operating conditions and column dimensions may be

deficient for understanding cluster size, which has a more direct correlation with the local conditions [36]. This underscores the importance of evaluating cluster characteristics based on local variables to mitigate contradictory results and improve the mechanistic understanding of the clustering phenomenon.

- **Particle properties**

Cluster size is affected by particle properties (e.g., size, polydispersity, particle size, surface roughness, etc), although specifications on the PSD of the particles investigated and reports on the impact of the type of polydispersity on cluster sizes have been limited. Previous studies have found that cluster size increases with particle size [21, 32, 50, 131] under riser flow conditions, especially as clusters of larger particles have higher cluster voidage [131]. Interestingly, high-speed video camera images found that clusters always consist of fine particles, such that no cluster was made up solely of large particles [2]. However, that study was performed in a fluidized bed, wherein a higher mass fraction of fines encouraged cluster formation and decreased entrainment rate [2], presumably due to higher inter-particle collisions and momentum transfers. Continuous PSD particles have been found to promote particle clustering compared to monodisperse particles [43, 65], so some mismatch in results may be due to different cluster characteristics in different flow regimes (e.g., dense fluidized bed, CFB riser or downer), and the reporting of a characteristic diameter rather than the actual PSD.

Table 4.3 lists the details of the particles reviewed in Fig. 4.2a, which shows that both Kiani et al. [131] and Xu and Zhu [32] reported similar cluster sizes at similar solid loading (m) conditions, despite the differences in their particle types (Table 4.3). Specifically, Kiani et al. [131] and Xu and Zhu [32] respectively investigated Geldart Groups A and B particles. Such a phenomenon could be due to greater wall effects in the small columns investigated in both studies, which are known to destroy clusters or

limit cluster growth [32, 131]. The wall effect has been observed to confer a narrower cluster size distribution for Geldart Group B particles was found near the wall [131].

Table 4.3. Tabulation of particle properties investigated in previous studies.

Year	Author(s)	Geldart Group	Material	ρ_p (kg/m ³)	d_p (μ m)
2000	Sharma et al. [21]	A	Glass beads	n/a	70
		B	Glass beads	n/a	120
2011	Chew et al. [53]	B	(i) Polystyrene (PS)	1050	327.5
		B	(ii) Glass beads	2500	165
		B	(iii) Sands	2650	196
2011 / 2012	Xu and Zhu [30, 32]	A	(i) FCC	1877	67
		A	(ii) Glass beads	2453	76
		B	(i) Glass beads	2403	134
		B	(ii) Glass beads	2498	288
		B	(iii) Sands	2467	138
2012	Chew et al. [44, 45]	B	(i) HDPE	900	650
		B	(ii) Glass beads	2500	170
		B	(iii) Glass beads	2500	650
2013	Kiani et al. [131]	B	(i) Glass beads	2640	150
		B	(ii) Glass beads	2640	300

*n/a = not available

Moreover, although Li et al. [41] and Wei et al. [129] examined particles with the same d_p of 54 μm (Table 4.1), Fig. 4.2b shows that the former gave cluster sizes of up to two orders-of-magnitude larger, even at approximately similar m conditions (e.g., $m = 17.9$ in Li et al. [41] and $m = 20.3$ in Wei et al. [129]) and axial positions, which could be due to the effect of particle density (ρ_p). Regarding ρ_p , Yin et al. [144] (not included in Fig. 4.2) has indicated that the relationship between ρ_p and clustering may not be monotonic, since higher ρ_p increases inter-particle collision but decreases viscous dissipation of gas flow, both effects of which influence cluster formation. As for surface properties, particles with lower coefficients of restitution was found to form larger clusters [99, 103], while particle surface conductivity was found to have negligible impact on particle clustering [99].

- **Correlations on cluster size**

Correlations have been developed for cluster size, as expressed as an equivalent diameter (d_c), per displayed in Table 4.4. Comparisons among predictions given by the correlations are not given here because the range of parameters used necessitates the substitution of too many values, a few of which are difficult to quantify experimentally, that may end up invalidating a fair comparison.

Table 4.4. Available correlations of cluster equivalent diameter (d_c).

Year	Author(s)	Correlation	Remarks
1986	Subbarao [101]	$d_c = \left[\frac{G_s}{U_g \rho_p (1 - \varepsilon_c)} \right]^{1/3} d_b$	Theoretical
1988	Horio et al. [145]	$d_c = \frac{\rho_g (U_{dil,g} - U_g)}{3g(\rho_p - \rho_g)(1 - \varepsilon_c)\varepsilon_g^{2(n-1)}}$	Semi-empirical

1994	Zou et al. [42]	$d_c = d_p \left[1.8543 \left(\frac{\varepsilon_g^{-1.5} (1 - \varepsilon_g)^{0.25}}{(\varepsilon_g - \varepsilon_{g,mf})^{2.41}} \right)^{1.3889} + 1 \right]$ <p>Limitation: Geldart Group A; $1.31 < U_g < 3.49$ (m/s); and $9.25 < G_s < 64.65$ (kg/m²/s)</p>	Semi-empirical
1994	Li and Kwauk [146]	$d_c = \frac{gd_p \left[\frac{U_p}{1 - \varepsilon_{max}} - \left(U_{mf} + \frac{\varepsilon_{c,mf} U_p}{1 - \varepsilon_{c,mf}} \right) \right]}{\frac{N_{st} \rho_p}{\rho_p - \rho_g} - g \left(U_{mf} + \frac{\varepsilon_{c,mf} U_p}{1 - \varepsilon_{c,mf}} \right)}$	Simulation (EMMS)
1999	Li et al. [147]	$d_c = \frac{d_p \left[\frac{f_{den} U_{den,s}}{1 - \varepsilon_{max}} - \left(U_{mf} + \frac{f_{den} \varepsilon_{c,mf} U_{den,s}}{1 - \varepsilon_{c,mf}} \right) \right]}{(f_{dil} U_{dil,g} - f_{den} U_{den,s}) - \left(U_{mf} + \frac{f_{den} \varepsilon_{c,mf} U_{den,s}}{1 - \varepsilon_{c,mf}} \right)}$	Simulation (EMMS)
1999	Xu and Kato [148]	$d_c = Ad_p \left(\frac{\rho_p}{\rho_c} \right)$ <p>Limitation: Geldart Groups A and B; $0.5 < U_g < 9$ (m/s); $20 < G_s < 120$ (kg/m²/s)</p>	Semi-empirical
2002	Harris et al. [149]	$d_c = \frac{\bar{\varepsilon}_g}{40.8 - 94.5 \bar{\varepsilon}_g}$ <p>Limitation: $54 < d_p < 310$ (μm); $930 < \rho_p < 3300$ (kg/m³); $1.2 < U_g < 4.8$ (m/s); $11.7 < G_s < 181$ (kg/m²/s); $0.05 < D < 0.65$ (m); $2.8 < H < 15.6$ (m)</p>	Semi-empirical
2004	Mostoufi and	<p>Ascending clusters:</p> $d_c = \exp[-0.47 + 1.276(U_g - U_{mf})]$	Semi-empirical

	Chaouki [150]	<p>Descending clusters:</p> $d_c = \exp[-1.79 + 1.555(U_g - U_{mf})]$ <p>Limitation: $70 < d_p < 385$ (μm); $1670 < \rho_p < 2650$ (kg/m^3); $0.5 < U_g < 2.8$ (m/s)</p>	
2005	Teplitskii, Y. S. [68]	$d_c = 40.4 H Fr \frac{\rho_g}{\rho_p}$, for $\frac{y}{H} \geq 0.2$	Semi-empirical

- **Remarks on discrepancies**

Three aspects that make the quantification of cluster sizes non-trivial and fraught with discrepancies among various reports are highlighted here. First, cluster sizes are known to vary spatially and temporally [5, 6, 139, 151], and are also affected by riser dimensions [21, 43, 131, 133], hydrodynamics (e.g., bubbles and jets [125], gas flow direction [66]), operating conditions (e.g., pressure [3], solid flux, and superficial gas velocity [11]), and particle properties (e.g., cohesiveness [152], particle diameter [125]); hence, the inevitable limited scopes of experiments (e.g., gas and solid flux ranges, riser locations, etc.) results in trends that may not be generally applicable. Second, another source of discrepancies among experimental results lies in the measurement being affected by (i) resolution, penetration depth, sensitivity, and width of the measurement area of the measurement instrument, (ii) threshold types (e.g., a fixed time-independent value [50] or time-dependent value [52]) and threshold values (minimum particle amount inside cluster, e.g., 6 [66, 69] or 100 [25, 47] or 1500 particles [2, 6, 36, 38, 153-156]), and (iii) type (e.g. chaos analysis, wavelet analysis) of the analysis methods. Third, clusters are of irregular [4, 42, 141] or other non-spherical shapes [29, 157] (e.g., arch-

shape [6, 11, 30, 31, 58, 66, 69, 125, 138, 139, 158-162] or stream-like [2, 9, 11, 42, 64, 163, 164]), hence the definition and/or measurement of a ‘characteristic’ size may differ.

4.3.2. Cluster duration

Some of the available publications expressed cluster size as cluster ‘duration’ [21, 32, 44, 45, 47, 53, 129], which was quantified by the continuous number of time instances whereby a higher concentration was detected by, for example the fiber optic probe, and expressed in unit of second. Cluster duration at various axial and lateral positions, and/or a broad range of polydispersity, particle properties, and operating conditions were found within the same study (see Table 4.1) are shown in Fig. 4.3 and Figs. 4.4 – 4.6 respectively for Geldart Groups A and B particles.

Fig. 4.3. Lateral distribution of cluster duration of monodisperse Geldart Group A

particles at (a) $h/H \approx 0.1 - 0.2$, (b) $h/H \approx 0.4 - 0.5$, and (c) $h/H \approx 0.6 - 0.7$.

• Lateral and axial trends

General observations are worth mentioning about the cluster duration trends of monodisperse particles for Geldart Groups A (Fig. 4.3) and B (Figs. 4.4a – c), both of which exhibit similar cluster duration magnitudes and trends. Cluster duration decreased with axial position and the cluster duration tended to increase as r/R or w/W increased lower in the riser (Fig. 4.3a and Fig. 4.4a). This was more significant with respect to r/R or w/W in the middle of the riser (Fig. 4.3b and Fig. 4.4b), but becomes negligible with respect to r/R or w/W higher in the riser (Fig. 4.3c and Fig. 4.4c). Furthermore, while the axial variations span up to two orders-of-magnitude, the lateral variations are lesser at up to one order-of-magnitude.

Despite cluster duration (Fig. 4.3) implying cluster size (Fig. 4.2), the former lacks the velocity component needed to make the data system-independent, hence even the qualitative comparison of the trends in Fig. 4.3 and Fig. 4.2 shows some contrasts, most notably in terms of the greater axial variations of cluster duration (Fig. 4.3). The difference could either be due to varying velocities axially, or the inaptness in comparing across different studies as noted earlier. Regarding the latter, it should be highlighted that Fig. 4.2 represents data mainly from Yang and Leu [47], whereas Fig. 4.3b – c are dominated by the data of Wei et al. [129] and Xu and Zhu [30]. Firstly, recall that Wei et al. [129] and Xu and Zhu [32], which used similar particle types, operating conditions, column configurations, and analysis equipment as an earlier work by the same authors (presented in Fig. 4.2) [30], also predicted lower cluster size than other experimental results in Fig. 4.2, which could be due to the intrusive nature of the probe in Wei et al. [129] and the dominant wall effect in the two-dimensional column in Xu and Zhu [30, 32]. Secondly, Fig. 4.3a presented data at much lower solid loading (m) conditions than that in Figs. 4.3b and c. Specifically, Yang and Leu [47] (Fig. 4.3a) characterized cluster duration spanning turbulent and fast fluidization regimes in the dense bed and splash

zones, while the other studies [21, 30, 129] in Figs. 4.3b – c assessed clusters in the freeboard of fast fluidization and/or dilute transport regimes.

- **Operating conditions**

Operating conditions (e.g., superficial gas velocity and solid mass flux) affect cluster formation-deformation and hence cluster duration. The relationship between cluster duration of monodisperse Group A particles and m shown in Fig. 4.3 is not clear, which is similar with the observations for cluster size (Fig. 4.2). In the case of Yang and Leu [47] (Fig. 4.3a) and Xu and Zhu [30] (denoted ‘i’ in Fig. 4.3c), both of which used FCC particles with similar particle diameter (d_p) and particle density (ρ_p) (Table 4.1), cluster duration decreased with m . On the other hand, similarly for FCC particles, Wei et al. [129] presented contrary trends in Fig. 4.3b in that cluster duration increased with m especially closer to the wall. Another pair of data from Xu and Zhu [30] (denoted ‘ii, $m = 15.4$ ’ in Fig. 4.3c) and Sharma et al. [21] ($m = 15.9$; Fig. 4.3b), both of which evaluated the same particle type (i.e., glass beads, $d_p = 70 - 76 \mu\text{m}$), is such that the latter gave cluster durations twice that of the former, indicating the lack of correlation between cluster duration and m . Fig. 4.4 shows the trends for Geldart Group B particles. Specifically, Fig. 4.4 contains nine sub-plots of cluster duration versus reactor height (h/H) to showcase the impact of solid loading (m) on the cluster duration of glass beads with particle diameters (d_p) in the range of $120 - 170 \mu\text{m}$ with varying extents of polydispersity [21, 30, 44, 45, 53]. The operating conditions evaluated in Fig. 4.4 are listed in Table 4.2

Table . Figs. 4.4a – c contain axial trends at three radial positions for glass beads with $d_p = 120 - 170 \mu\text{m}$ for $m = 0.1 - 30.9$ [21, 30, 44, 45, 53], Figs. 4.4d – f contain axial trends at three radial positions for a binary size mixture (namely, 50 – 50 vol. %

of glass beads with $d_p = 170 \mu\text{m}$ and $650 \mu\text{m}$) for $m = 5.9 - 16$ [44, 45], while Figs. 4.4g – i contain axial trends at three radial positions for a Gaussian PSD (with $\sigma/d_p = 0.25$) of glass beads with $d_p = 170 \mu\text{m}$ for a solid loading (m) range of $5.9 - 25$ [44, 45].

Fig. 4.4. The impact of solid loading on cluster duration for Geldart Group B particles of (a – c) monodisperse glass beads with $d_p = 120 - 170 \mu\text{m}$, (d – f) binary size

mixture (i.e., 50 – 50 vol. % of glass beads with $d_p = 170\ \mu\text{m}$ and $650\ \mu\text{m}$), and (g – i) Gaussian PSD with $\sigma/d_p = 0.25$ of glass beads with $d_p = 170\ \mu\text{m}$.

Figs. 4.4a – c, which contain data from different studies on similar particle type and diameter (i.e., glass beads with $d_p = 120 - 170\ \mu\text{m}$ [21, 30, 44, 45, 53]), show similarly as for Geldart Group A particles of the lack of a relationship between cluster duration and m for the monodisperse Geldart Group B particles. Even among the data from the same study (i.e., same CFB riser, instrument, and analysis method), Fig. 4.4a – i show no clear correlation between cluster duration and m . Two key observations are noted here. Firstly, cluster duration spans the same range of values (i.e., $0.01 - 0.1\ \text{s}$) despite the differences in axial positions, operating conditions, and polydispersity types. The much smaller cluster durations ($< 0.01\ \text{s}$) of Xu and Zhu [30] (Fig. 4.4c) is presumably due to the higher wall friction in the two-dimensional rectangular column investigated [32, 131], although the axial position was also slightly lower ($h/H \approx 0.7$) and particle diameter slightly smaller ($d_p = 134\ \mu\text{m}$). Secondly, the cluster duration of glass beads with $d_p = 120 - 170\ \mu\text{m}$ generally increases laterally (from the center to the wall) lower in the riser, but the lateral profile becomes flatter with riser height.

- **Polydispersity**

The contradictory results among previous experimental data are so at least in part due to incomplete data sets and different methods (i.e., both in terms of measurement and analysis), which thereby hinder a mechanistic understanding of cluster characteristics. Therefore, a compilation of comprehensive experimental datasets via the same measurement technique and analysis method in Figs. 4.4a – i [44, 45, 53] will be re-plotted next, in order to improve the readability of the graph and provide a more

complete landscape, at least qualitatively, on the impact of polydispersity throughout the riser.

Fig. 4.5 summarizes experimental data for glass beads involved in three types of polydispersities, namely, monodisperse glass beads with $d_p = 170 \mu\text{m}$, binary size (i.e., two species with the same ρ_p but different d_p ; in this case, glass beads with $d_p = 170 \mu\text{m}$ and $650 \mu\text{m}$) and continuous PSD (i.e., a Gaussian PSD, with a ratio of standard deviation to mean diameter (σ/d_p) of 0.25) glass beads with $d_p = 170 \mu\text{m}$, for lower and higher solid loadings (m) at various axial and radial positions in the riser.

Fig. 4.5. The impact of polydispersity on cluster duration of Geldart Group B particles at (a – c) lower solid loading ($m = 4.2 - 5.9$), and (d – f) higher solid loading ($m = 16.0 - 16.7$).

Specifically, each of the six sub-plots in Fig. 4.5 represents cluster duration as a function of dimensionless height (h/H , where h is the height evaluated and H is the total height of the riser) at three dimensionless radial positions (r/R , where r is the radial position evaluated and R is the total radius of the riser); the upper row of three sub-plots (Figs. 4.5a – c) represent the lower solid loading condition, m , of 4.2 – 5.9, while the lower row (Figs. 4.5d – e) represent the higher m values of 16.0 – 16.7 [44, 45]. The ranges of the x - and y -axes are kept the same for all six sub-plots for easier comparison. Previous section also found that the cluster duration of monodisperse Geldart Groups A and B span largely the same range (see Fig. 4.3 and Fig. 4.4).

The more complete dataset in Fig. 4.5 sheds some light on the contradictory trends noted earlier. With respect to axial trends, the most significant axial variations were displayed by the monodisperse system at the wall (Figs. 4.5c and f). In the riser center (i.e., between $r/R = 0$ and 0.5), cluster duration tended to be either increasing slightly or be invariant with riser height (h/H) for all three types of polydispersities and at both m conditions. On the other hand, decreasing duration trends axially was observed for the continuous PSD at the lower m condition (Fig. 4.5c). Regarding radial trends, cluster duration increased radially for all three polydisperse types at the higher m condition (Figs. 4.5d – f) and only for the monodisperse type at the lower m condition (Figs. 4.5a – c). On the contrary, cluster duration exhibited decreasing radial trends at some heights for the binary mixture and continuous PSD at the lower m condition (Figs. 4.5a – c). Similar to previous observation for monodisperse particles [25, 36], cluster duration of polydisperse systems (i.e., binary and continuous PSDs) has wider range near wall compared to core area, which shows higher inter-particle and wall-particle interactions affect cluster growth. Furthermore, the monodisperse particle system has a wider cluster duration range compared to polydisperse system, which may be related to

the more pronounced inter-species momentum transfer and species segregation in the latter [126, 165].

Two observations in Fig. 4.5 on the impact of polydispersity on cluster duration should be noted: (i) throughout the riser and at both solid loading (m) conditions, the monodisperse particles exhibited the highest cluster duration while the continuous PSD system exhibited the lowest cluster duration; and (ii) the monodisperse particles displayed the greatest axial variations at the wall ($r/R = 0.94$) and at both m conditions (Figs. 4.4c and f). The discrepancies in the cluster duration trends may lie in momentum transfer and species segregation associated with particles with either different particle size (d_p), particle density (ρ_p) or particle type [126, 166], both of which are unique to non-monodisperse systems. Regarding (i), the highest cluster duration given by the monodisperse system may be because the lack of inter-species momentum transfer served to mitigate cluster breakup, while the widest range of inter-species momentum transfer values in the continuous PSD system served to inhibit cluster growth. Particularly at the wall ($r/R = 0.94$), due to the preferential size segregation of larger particles to the wall only for the binary size and continuous PSD systems [126, 165], the cluster durations for these two systems were depressed relative to that for monodisperse. As Fig. 4.5f shows, the larger glass beads ($d_p = 650 \mu\text{m}$) exhibited lower cluster durations than smaller glass beads ($d_p = 170 \mu\text{m}$) at $r/R = 0.94$.

Concerning other types of polydispersity, the same study pointed out that the cluster duration of the binary density mixture (i.e., 50 – 50 vol. % of glass beads and HDPE, both with $d_p = 650 \mu\text{m}$) was lesser than that of binary size (i.e., 50 – 50 vol. % of glass beads with $d_p = 170 \mu\text{m}$ and $650 \mu\text{m}$) throughout the riser, and the former displayed a lower cluster duration compared to its monodisperse constituents while the latter mimicked the cluster duration of the larger monodisperse constituent [45]. As for

the impact of PSD width of continuous PSD particles and mixture composition of binary mixtures of Geldart Group B particles, cluster duration only differed among the systems at the bottom of the dilute CFB riser [45, 53].

- **Particle properties**

In the previous section on cluster size, no clear, consistent trend was found between cluster size and d_p or ρ_p . Previous studies have found that cluster size (and hence cluster duration) could increase with d_p [21, 30, 50, 131] in a CFB riser. This is in contrast with the decrease with d_p in a fluidized bed [2]. Different regimes likely give rise to different clustering mechanisms. Similarly for cluster duration, (i) it was also noted that the cluster duration of Geldart Groups A and B span the same range (Fig. 4.3 versus Figs. 4.4a – c), and (ii) Fig. 4.3c shows that the experimental results of Xu and Zhu [30], with particles of similar d_p (67 – 76 μm) but different ρ_p (denoted ‘i’ and ‘ii’ in the legend, which correspond respectively to $\rho_p = 1877$ and 2453 kg/m^3 (see Table 4.3) indicate that a higher ρ_p caused negligible impact on cluster duration at a lower m (15.4) but a higher cluster duration at a higher m (21.2).

The impact of particle properties has been reported to be less significant than operating conditions and riser position [44]. In order to more clearly understand the impact of particle size and density, Fig. 4.6 shows six plots of cluster duration versus h/H of various monodisperse particles (Table 4.3) at lower [53] and higher [44] solid loadings (m). For the lower m condition (Figs. 4.6a – c), the three monodisperse particle species displayed are sand, glass beads with $d_p = 165 \mu\text{m}$, and polystyrene (PS); for the higher m condition, the four monodisperse particle species displayed are sand, small glass beads ($d_p = 120 - 288 \mu\text{m}$), large glass beads ($d_p = 650 \mu\text{m}$), and high-density polyethylene (HDPE).

Fig. 4.6. The impact of particle properties on cluster duration of Geldart Group B particles at (a – c) lower solid loading ($m = 0.1 - 0.2$), and (d – f) higher solid loading ($m = 15.4 - 16.0$).

Fig. 4.6 shows that the impact of particle property on cluster duration (i.e., the differences among the trends) was as significant as that of polydispersity type (Fig. 4.5), which underscores the need for considering polydispersity. Two notes on the impact of d_p are worth highlighting: (i) the comparison between Chew et al. [44] ($d_p = 170 \mu\text{m}$, $m = 16.0$) and Sharma et al. [21] ($d_p = 120 \mu\text{m}$, $m = 15.9$), both which used glass beads at similar solid loading condition (m), shows in Fig. 4.6e that a smaller d_p gave a significantly lower cluster duration at $r/R = 0 - 0.5$, and (ii) the comparison

between the glass beads results of Chew et al. [44] ($d_p = 170\ \mu\text{m}$, $m = 16.0$) and Xu and Zhu [30] ($d_p = 134\ \mu\text{m}$, $m = 15.4$) in Fig. 4.6f indicated that a slightly lower d_p resulted in a decrease in cluster duration by one order-of-magnitude, and the impact of d_p between the glass beads results within the same study was also not consistent. Recall that the different riser configurations and cluster analysis methods used in different studies affect the cluster behavior results obtained. Regarding (i), although Sharma et al. [21] and Chew et al. [44] used similar riser diameters, Sharma et al. [21] used a capacitance probe whereas Chew et al. [44] used a fiber optic probe. As for (ii), while Xu and Zhu [30] used a two-dimensional riser with a rectangular cross-section, Chew et al. [44] used a three-dimensional riser with a circular cross-section. Furthermore, the data acquisition frequency was two orders-of-magnitude smaller in Chew et al. [44], although still well above the characteristic 10 Hz of clusters. The quantitative comparison on the impact of particle properties and operating condition among different studies should be done cautiously.

To attempt to shed some light on the contradictory reports of the impact of particle diameter (d_p) on clustering, the studies involving various d_p values of glass beads (see Table 4.3) were inspected in more detail. Sharma et al. [21] investigated glass beads with $d_p = 70\ \mu\text{m}$ (Group A, Fig. 4.3) and $120\ \mu\text{m}$ (Group B, Fig. 4.4), and found that a higher d_p gave a higher cluster duration at a lower solid loading ($m = 9.7$) but the impact of d_p was negligible at a higher solid loading ($m = 15.9$). Xu and Zhu [30] studied a wider d_p range of glass beads (namely, $d_p = 76\ \mu\text{m}$ (Group A, Fig. 4.3), $d_p = 134\ \mu\text{m}$ (Fig. 4.6f), and $d_p = 288\ \mu\text{m}$ (Fig. 4.6f)), and found similarly that a larger d_p gave a larger cluster duration at $m = 15.4$, but the impact of d_p was negligible at a higher solid loading ($m = 30.9$). Chew et al. [44] further probed the d_p effect with a larger d_p differential of 170 and 650 μm , and found in contrast that the larger d_p gave a lower cluster duration at

the lower m (5.9 – 7.4), and also that a higher d_p gave a higher cluster duration at the riser core but lower near the wall at the highest m ($m = 16.0$; Figs. 4.6e – f). Collectively, it appears that the impact of d_p on clustering is not so straightforward but rather dependent on an interplay of riser configuration and operating conditions.

4.3.3. Cluster frequency

Similar to cluster size and duration, cluster frequency has been reported to both increase [36] and decrease [43, 167] with respect to radial position (r/R), and also both increase and decrease with respect to axial positions (h/H) [66, 168] have been reported. Fig. 4.7 compiles previous experimental results on the radial profile of cluster frequency for monodisperse Geldart Group A particles.

Fig. 4.7. Lateral distribution of cluster frequency of Geldart Group A particles at (a)

$h/H \approx 0.2 - 0.3$, (b) $h/H \approx 0.6 - 0.7$, and (c) $h/H \approx 0.8 - 0.9$.

- **Lateral and axial trends**

Fig. 4.7 displays the lateral cluster frequency profiles of monodisperse Geldart Group A particles under various solid loading (m) conditions and riser axial heights. As seen in Fig. 4.7, the cluster frequency has been found to span two orders-of-magnitude, and to either increase, decrease, or be approximately invariant as r/R or w/W or h/H increased. The lack of a clear trend between cluster frequency and riser position agrees with that of Group B particles shown in Fig. 4.8. Analogous to Fig. 4.7, Fig. 4.8 presents the lateral cluster frequency profiles of monodisperse (Figs. 4.8a - c), binary (Figs. 4.8d - f), and continuous PSD (Figs. 4.8g - i) of Geldart Group B particles. The key difference between the lateral cluster frequency profiles of Geldart Groups A (Fig. 4.7) and B (Fig. 4.8) particles is a wider range of cluster frequency values was found for the former, which is presumably due to the wider range of solid loading (m) conditions studied.

- **Operating conditions**

Higher solid concentration has been shown to lead to higher growth rate coefficients in perturbation waves and lower particle stability, both of which encourage cluster formation [47, 56, 65] and cluster frequency [130, 142]; but more clusters have also been found in more dilute regions [11, 169], as substantiated by a simulation effort indicating that the cluster disappearance rate near the wall was higher than in the center [65]. Yet another experimental effort showed that cluster frequency was non-monotonically (i.e., increased then decreased) correlated with solid loading and local solid concentration [36]. Such contradictory trends may be related to the decreasing and increasing trends of cluster frequency respectively for smaller (i.e., corresponding to cluster duration < 15 ms) and bigger clusters as local solid concentration increased [36].

With respect to superficial gas velocity, a study showed that cluster frequency decreased for larger particles and remained relatively constant for smaller particles [21,

142]. Other experimental and simulation efforts indicated that cluster formation was lesser at higher superficial gas velocities [23, 47, 56, 129, 170], while others have reported that higher superficial gas velocities did not form nor destroy clusters, but simply increased cluster voidage [171, 172]. A simulation effort on a dilute two-dimensional rectangular column found no clear trend between both local superficial gas velocity and solid concentration on cluster formation-deformation [71]. Another effort further suggested an upper Reynolds number (Re_p) limit of 1800, beyond which a cluster of spherical particles became unstable [154]. It was also proposed that the use of superficial gas velocity and solid loading to evaluate clustering phenomena may not be relevant because, even for the same superficial gas velocity, the local superficial gas velocity in a dense system can be much higher than that of dilute one [169]; hence, the evaluation of clustering based on localized conditions would serve to reduce contradictions among results of various studies and lead to a more fundamental understanding of the clustering phenomenon.

Both Fig. 4.7 and Fig. 4.8 show the lack of a clear relationship between cluster frequency and solid loading (m) respectively for Geldart Groups A and B particles, which is similar to the observation for cluster duration. Two highlights regarding the impact of m on cluster frequency should be noted: (i) even for the data obtained within the same study (e.g., Manyele et al. [36]), the correlation between cluster frequency and m was non-monotonic; and (ii) the cluster frequency trends tended to be closer to one another if they belong to the same study, which perhaps indicate the more dominant influence of riser configuration, instrument used, or analysis method employed compared to m .

Fig. 4.8. The impact of solid loading on cluster frequency for Geldart Group B particles of (a – c) monodisperse glass beads with $d_p = 120 - 170 \mu\text{m}$, (d – f) binary size mixture (i.e., 50 – 50 vol. % glass beads with $d_p = 170 \mu\text{m}$ and $650 \mu\text{m}$), and (g – i) continuous (Gaussian) PSD with $\sigma/d_p = 0.25$ of glass beads with $d_p = 170 \mu\text{m}$ particles.

- **Polydispersity**

Fig. 4.9 shows that the cluster frequency trends of glass beads of three types of polydispersities, namely, monodisperse ($d_p = 120 - 170 \mu\text{m}$), binary size mixture of 50 – 50 vol. % ($d_p = 170 \mu\text{m}$ and $650 \mu\text{m}$), and Gaussian PSD with $\sigma/d_p = 0.25$ and $d_p = 170 \mu\text{m}$. Specifically, Fig. 4.9 contains six sub-plots of cluster frequency versus r/R , with the first row (Figs. 4.9a – c) representing the lower m condition at three axial positions, and the second row (Figs. 4.9d – f) representing the higher m condition. The ranges of the x - and y -axes are kept the same for all sub-plots for easier comparison. The range of cluster frequency in Fig. 4.9 is approximately 5 – 20 Hz, which encompasses the range of 6 – 11 Hz reported in a previous simulation effort [21]. Three observations are worth noting: (i) compared to the monodisperse and continuous PSD systems, the binary size mixture generally displayed the lowest cluster frequency throughout the riser and at both m conditions; (ii) the monodisperse and continuous PSD systems exhibited similar cluster frequency values throughout the riser and at both m conditions; and (iii) unlike cluster duration, cluster frequency ranges of various polydispersity systems are largely similar near wall and core area. Regarding (ii), this implies that the inter-particle momentum transfer effects among the disparate species in the binary size mixture of glass beads with $d_p = 170 \mu\text{m}$ and $650 \mu\text{m}$ appeared to decrease cluster frequency, regardless of solid loading or riser position. On the other hand, although the continuous PSD system also necessitated inter-species momentum transfer among the different size species, the effect was lesser such that it behaved similar to the monodisperse system with regards to cluster frequency. Recalling from Fig. 4.5 that the monodisperse particles exhibited the highest cluster duration while the continuous PSD system exhibited the lowest cluster duration, the effect of inter-species momentum transfer in the various types of polydisperse systems on cluster frequency appears different from

that on cluster duration. Furthermore, (iii) indicates that wall friction affects cluster growth more than cluster formation/deformation.

Fig. 4.9. The impact of polydispersity on cluster frequency of Geldart Group B particles at (a – c) lower solid loading ($m = 4.2 - 5.9$) and (d – f) higher solid loading ($m = 16.0 - 16.7$).

• Particle properties

Previous experimental efforts using Geldart Groups A and B particles in two-dimensional columns indicated that particle diameter and density had a greater effect on cluster formation than particle shape, in particular that smaller particles had a higher tendency to form clusters due to stronger inter-particle forces [30, 43]. Larger particles, which formed larger clusters [21, 30, 131], had been shown to reduce the number of

clusters in the system [36]. On the cluster frequency of Geldart Group A particles (Fig.), Guenther and Breault [38] constantly gave higher cluster frequency than other studies, even when compared to the studies with similar solid loading (m) [47, 130]. Recall that Guenther and Breault [38] tends to give the largest cluster size (Fig. 4.2) and highest cluster frequency (Fig. 4.8), presumably due to the use of cork, whose d_p (812 μm) and ρ_p (189 kg/m^3) were considerably larger and smaller, respectively, compared to the more popularly studied Geldart Group A particles (e.g., FCC).

Fig. 4.10 shows the impact of particle properties (i.e., both particle diameter and density) of monodisperse Geldart Group B particles on cluster frequency [30, 44, 53]. The top row of three sub-plots (Figs. 4.10a – c) display the cluster frequency trends at three radial positions for monodisperse glass beads with $d_p = 165 \mu\text{m}$, sand, and PS at the lower m condition of 0.1 – 0.2, while the bottom row of three sub-plots (Figs. 4.10d – f) display the cluster frequency trends at three radial positions for monodisperse small glass beads ($d_p = 134 - 288 \mu\text{m}$), large glass beads ($d_p = 650 \mu\text{m}$), and HDPE at the higher m condition ($m = 15.4 - 16$).

Fig. 4.10. The impact of particle properties on cluster frequency of Geldart Group B particles at (a – c) lower solid loading ($m = 0.1 - 0.2$) and (d – f) higher solid loading ($m = 15.4 - 16.0$).

With respect to cluster frequency, two key highlights regarding the impact of particle properties are noted: (i) sand, which was the least spherical compared to glass beads with $d_p = 165 \mu\text{m}$ and PS at the lower m condition, exhibited the highest cluster frequency (Figs. 4.10a – c), which could be due to the increased particle rotation leading to enhanced collisional cooling and thereby the formation of more clusters [2]; (ii) within the same study [44], large glass beads ($d_p = 650 \mu\text{m}$), whose Stokes number (St) was the greatest compared to small glass beads ($d_p = 170 \mu\text{m}$) and HDPE at the higher m condition, exhibited the lowest cluster frequency at the riser core ($r/R = 0$; Figs. 4.10e

– f), but highest near the wall ($r/R = 0.94$; Figs. 4.10e – f); and (iii) in spite of differences in both particle diameter and density, glass beads with $d_p = 165 \mu\text{m}$ and PS gave similar cluster frequencies at lower m (Figs. 4.10a – c), while glass beads with $d_p = 170 \mu\text{m}$ and HDPE gave largely similar cluster frequencies at higher m (Figs. 4.10d – f). Observations (ii) and (iii) are similar to those made for cluster duration (Fig. 4.6). Regarding (ii), the physical picture could be that, while the high St associated with large glass beads ($d_p = 650 \mu\text{m}$) made it more resistant to cluster breakup (hence higher cluster duration and lower cluster frequency) in the more dilute riser core ($r/R = 0$; Figs. 4.10e – f), it also made it more prone to cluster breakup (hence lower cluster duration and higher cluster frequency) in the higher concentration of large glass beads ($d_p = 650 \mu\text{m}$) particles near the wall ($r/R = 0.94$; Figs. 4.10e – f). Observation (iii) agrees with the cluster duration trends in terms of riser position being a more dominating influence [44, 45, 53] on clusters relative to the disparities in particle properties between the two species.

4.3.4. Cluster appearance probability

Similar to the previous sections on cluster duration and cluster frequency, the following sections evaluate more comprehensive dataset to understand the impact of various parameters on cluster appearance probability.

- **Lateral and axial trends**

Experimentally, studies of Geldart Group A particles showed constant [47] or decreasing trends as r/R increased [38], while Geldart Group B particles showed increasing trend as r/R increased [48]. In parallel, simulations showed increasing [4, 36, 65] and decreasing trends [56] for Geldart Groups A and B particles, respectively, with respect to r/R . With respect to axial variations, experimental results have shown that the

appearance probability of clusters decreased [2], increased [23, 36, 48, 129], or remained constant [47] with height. Riser-wide studies using Geldart Group B particles in a dilute riser [53] found no clear correlation between cluster appearance probability and axial position for monodisperse and binary mixtures, while general increasing trend axially was found for continuous PSD particles. In order to understand the seemingly contradictory trends of cluster appearance probability in the CFB riser, Fig. 4.11 compares the lateral profiles of cluster appearance probability from previous experimental results of monodisperse Geldart Group A [129] particles at $h/H \approx 0.5$.

Fig. 4.11. Lateral distribution of cluster appearance probability at $h/H \approx 0.5$ for Geldart Group A particle [129].

Two interesting observations in Fig. 4.11 are noted. Firstly, the magnitudes of cluster appearance probability of Group A particles (Fig. 4.11) were significantly lower than those of Group B (Figs. 4.12a – c). The similarity between the two studies is that both used intrusive probes, but the m conditions investigated for Group A were much higher than those for Group B. Secondly, whereas the Geldart Group A particles exhibited a distinct increase in cluster appearance probability as r/R increased (Fig. 4.11), that of Group B appeared to be non-monotonic with respect to r/R (Figs. 4.12a – c).

Regarding the radial trends, Fig. 4.11 illustrates that the cluster appearance probability tended to increase with r/R , particularly near the bottom of the riser, and increased with r/R near the top of the riser for the lower m conditions. Regarding the axial trends, the most significant axial variations are consistently at $r/R \approx 0.9$ (Fig. 4.12).

- **Operating conditions**

Fig. 4.12 illustrates the relationship between cluster appearance probability and m for various types of polydispersities involving glass beads with $d_p = 120 - 170 \mu\text{m}$. Specifically, Figs. 4.12a – c display the axial trends of monodisperse glass beads with $d_p = 120 - 170 \mu\text{m}$ at three different radial positions for $m = 0.1 - 16.0$, Figs. 4.12d – f display axial trends of the binary size mixture (i.e., 50 – 50 vol. % glass beads with $d_p = 170 \mu\text{m}$ and $650 \mu\text{m}$) for $m = 5.9 - 16.0$, while Figs. 4.12g – i display axial trends of the Gaussian PSD of glass beads with $d_p = 170 \mu\text{m}$ (with $\sigma/d_p = 0.25$) for $m = 5.9 - 25.0$.

Fig. 4.12. The impact of solid loading on cluster appearance probability for Geldart Group B particles of (a – c) monodisperse glass beads with $d_p = 120 - 170 \mu\text{m}$, (d – f) binary size mixture (i.e., 50 – 50 vol. % glass beads with $d_p = 170 \mu\text{m}$ and $650 \mu\text{m}$), and (g – i) continuous (Gaussian) PSD with $\sigma/d_p = 0.25$ of glass beads with $d_p = 170 \mu\text{m}$ particles.

The cluster appearance probability of Geldart Group A particles largely increased with m particularly closer to the wall (Fig. 4.11), but the positive correlation did not strictly hold for the Geldart Group B particles (Fig. 4.12). Fig. 4.12 affirms the lack of a consistent correlation between cluster appearance probability and m for Geldart Group B particles. Two key observations are noted. Firstly, at $r/R \approx 0.9$ (Fig. 4.12), while the higher m conditions tended to give higher cluster appearance probability at the bottom of the riser, the effect reversed at the top of the riser in that the higher m conditions tended to give lower cluster appearance probability. This implies that the effect of m on clustering varies at different axial positions of the riser, especially near the wall. Secondly, at $r/R \leq 0.5$, cluster appearance probability generally tended to decrease as m increased for all three particle systems. This contradicts the observation in Fig. 4.11 on Geldart Group A particles, possibly due to the higher m values investigated for the latter.

- **Polydispersity**

Fig. 4.13 shows the cluster appearance probability trends of three types of polydisperse systems involving glass beads with $d_p = 120 - 170 \mu\text{m}$, namely, monodisperse with $d_p = 170 \mu\text{m}$, binary size (i.e., 50 – 50 vol. % of $d_p = 170 \mu\text{m}$ and $650 \mu\text{m}$) and continuous (Gaussian) PSD with $\sigma/d_p = 0.25$ and $d_p = 170 \mu\text{m}$. Each of the three trend lines in the six sub-plots represent each polydisperse type. The first row of Fig. 4.13 contains three sub-plots representing three radial positions under the lower solid loading (m) condition of $m = 4.2 - 5.9$ (Figs. 4.13a – c). The second row is analogous to the first but at the higher m condition of $m = 16.0 - 16.7$ (Figs. 4.13d – f).

Fig. 4.13. The impact of polydispersity on cluster appearance probability of Geldart Group B particles at (a – c) lower solid loading ($m = 4.2 - 5.9$) and (d – f) higher solid loading ($m = 16.0 - 16.7$).

Three observations in Fig. 4.13 are worth highlighting regarding the impact of polydispersity on cluster appearance probability. Firstly, the variations in cluster appearance probability due to polydispersity type tended to be most significant at $r/R = 0.94$, which is consistent to that for both cluster duration (Fig. 4.5) and frequency (Fig. 4.9). This implies that the higher solid concentration in the annulus and/or the wall effect enhanced the impact of the differences in polydispersity type. Secondly, compared to the other two particle systems, the binary size mixture tended to give higher cluster appearance probability at $r/R \leq 0.5$, but lower cluster appearance probability at $r/R =$

0.94. Recall that, among the three types of polydispersity, the binary mixture gave intermediate cluster duration (Fig. 4.5) and the lowest cluster frequency (Fig. 4.9). Thirdly, for $r/R \leq 0.5$ and under both m conditions, whereas the monodisperse and continuous PSD particle systems exhibited similar magnitudes of cluster frequency (Fig. 4.9) and cluster appearance probability (Fig. 4.13), the magnitudes of cluster duration were distinctly different between the two particle systems. This indicates that polydispersity has different impact on cluster formation (associated with cluster frequency) and cluster growth (associated with cluster duration).

- **Particle properties**

Particles properties (i.e., particle size, density, surface properties, shape, porosity, etc) are well-acknowledged to affect cluster formation and deformation [44, 45, 53, 70, 105, 173, 174]. Fig. 4.14 illustrates the cluster appearance probability of five monodisperse Geldart Group B particles (Table 4.3) to attempt to understand the impact of d_p and ρ_p from the comprehensive dataset within the same study [44, 53]. The first and second rows show the axial trends at three radial positions respectively under the lower m condition of 0.1 – 0.2 (Figs. 4.14a – c) [53] and the higher m condition of 16.0 (Fig. d – f) [44]. Specifically, three types of monodisperse particles were evaluated at the lower m condition (Figs. 4.14a – c), namely, sand, glass beads with $d_p = 165 \mu\text{m}$, and polystyrene (PS), and three at the higher m condition (Figs. 4.14d – f), namely, glass beads with $d_p = 170 \mu\text{m}$, glass beads with $d_p = 650 \mu\text{m}$, and high-density polyethylene (HDPE).

Fig. 4.14. The impact of particle properties on cluster appearance probability of Geldart Group B particles at (a – c) lower solid loading ($m = 0.1 - 0.2$) and (d – f) higher solid loading ($m = 16.0$).

The comparison of the cluster appearance probability of various monodisperse particles in Fig. 4.14 leads to one key conclusion. Specifically, the impact of the particle properties of monodisperse particle systems on cluster appearance probability was surprisingly insignificant, which seems different to that observed for cluster duration (Fig. 4.6) and frequency (Fig. 4.10), whereby the magnitudes changed when the particle systems were different. This implies that particle properties exert different impact on cluster formation (associated with cluster frequency) and cluster growth (associated with

cluster duration), which is consistent with the earlier hypothesis regarding the effect of polydispersity type.

4.4. Conclusions

The non-negligible impact of particle clustering on the overall performance of gas-solid fluidization systems is well-acknowledged, which has led to the generation of abundant experimental and simulation studies on the clustering phenomenon in the past seven decades. This review aims at summarizing the experimental data on cluster characteristics in circulating fluidized bed (CFB) risers to provide an overview of the understanding to date. A synopsis of the experimental and simulation methods harnessed, and also the origin of clusters was first given. Then, the impact of various parameters (namely, local riser position, operating condition, particle property, polydispersity) on cluster characteristics (namely, size, duration, frequency, and appearance probability) was examined.

Collectively, the information available on clusters have progressed. Unfortunately, apparent contradictory results found in the literature serve to hamper rather than advance model validation, which in turn obstructs a mechanistic understanding of the clustering phenomenon. The discrepancies in the clustering trends can be traced to three key reasons: (1) cluster characteristics are affected by an interplay of a host of factors (e.g., riser configuration, riser local position, operating condition, particle property, among others), hence, the inevitable limited scopes of experiments (e.g., gas and solid flux ranges, riser locations, etc.) results that may not be generally applicable; (2) cluster characterization is affected by both the choice of instrument (e.g., invasive or non-invasive) and analysis method (e.g., wavelet analysis versus a simple averaging); and (3) clusters are of irregular shapes, hence the definition and/or

measurement of the various cluster characteristics (e.g., size) invariably differ. Hence, a compilation of the more comprehensive experimental datasets via the same measurement technique and analysis method from previous publications (Table 4.1) were presented, in order to provide a more complete landscape on the clustering phenomenon.

Three observations are worth noting. Firstly, the quantitative comparison of clusters across studies must be treated with caution, because discrepancies of an order-of-magnitude was observed by different studies of similar particles and operating conditions. Along the same lines, the cluster trends tended to be closer to one another if they belong to the same study, which perhaps indicate the more dominant influence of riser configuration, instrument used, or analysis method employed compared to the actual parameter investigated. Secondly, a straightforward correlation between any parameter (e.g., particle diameter) and any cluster characteristic (e.g., cluster duration) is not possible, presumably because of the interplay of multiple factors, such that the trend is unclear even within same study (e.g., the relationship between solid loading (m) and cluster frequency). Thirdly, in view of polydispersity having a non-negligible impact on clustering, some mismatch in results may well be due to the mere reporting of a characteristic diameter rather than the actual particle size distribution (PSD). The inter-species momentum transfer and species segregation effects in non-monodisperse systems are expected to affect both cluster formation (associated with cluster duration) and cluster growth (associated with cluster frequency).

What is remarkable is that the trends of the cluster characteristics are relatively consistent despite different experimentalists and units. The key contradiction in trends is due to the difference of flow regimes. Specifically, the clusters in a fluidized bed versus those in a CFB riser are expected to originate from different mechanisms.

Nomenclature

A	= constant of Xu and Kato [148] correlation, –
D	= riser diameter, m
d_c	= cluster diameter, μm
d_p	= particle diameter, μm
f_{den}	= volume fraction of particle-rich phase, –
f_{dil}	= volume fraction of gas-rich phase, –
Fr	= Froude number = $\frac{U_g}{\sqrt{gd_p}}$, –
g	= gravitational acceleration, m/s^2
G_s	= solid mass flux, $\text{kg/m}^2/\text{s}$
H	= riser height, m
m	= solid loading = $G_s/(\rho_g U_g)$, –
n	= parameter in Richardson and Zaki [199] correlation = $\frac{\ln(\frac{U_g}{U_t})}{\ln \varepsilon_g}$, –
N_{st}	= mass specific energy consumption of particles, W/kg
r	= radial position evaluated, m
R	= riser radius, m
Re_p	= Reynold number of particle = $\frac{\rho_g U_g d_p}{\mu_g}$, –
$U_{den,s}$	= particle velocity in particle-rich phase, m/s
$U_{dil,g}$	= superficial gas velocity in gas-rich phase, m/s
U_g	= superficial gas velocity, m/s
U_{mf}	= minimum fluidization velocity, m/s
U_p	= superficial particle velocity, m/s
w	= axial position evaluated, m
W	= riser width (measured from riser center), m

Symbols and Greek Letters

σ	= standard deviation, –
$\varepsilon_{c,mf}$	= cluster voidage at minimum fluidization velocity, –
ε_g	= system voidage, –
$\varepsilon_{g,mf}$	= system voidage at minimum fluidization velocity, –
ρ_c	= cluster density, kg/m^3
ρ_g	= gas density, kg/m^3
ρ_p	= particle density, kg/m^3

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CHAPTER 5: THE IMPACT OF MONODISPERSE GAC FLUIDIZATION ON MEMBRANE FOULING MITIGATION IN ANFMBR

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Abstract

The anaerobic fluidized bed membrane bioreactor (AnFMBR) utilizes the fluidization of granular activated carbon (GAC) particles as an unsteady-shear means for mitigating membrane fouling. In this study, a two-fluid model (TFM) was validated against experimental results and used to obtain a comprehensive landscape of local water and particle velocities and concentrations, which directly related to particle momentum and water shear stress as membrane fouling mitigation mechanism. The results provided interesting insights. Firstly, the larger particles conferred greater particle momentum (i.e., extent of fouling mitigation) at the same water shear stress magnitude, but at the cost of higher power of pump consumption. Smaller particles can be fluidized at relatively lower power of pump, but they provide smaller local particle momentum transfer. Secondly, at the same power input, the water shear stress or particle momentum values for the different particle diameters overlapped, which indicates that other parameters (e.g., superficial water velocity) can be manipulated to improve

performance even when particle diameter is fixed. It should be noted that monodisperse (i.e., single particle sizes) particles were studied here, but practical systems are inevitably polydisperse (i.e., range of particle sizes), so future studies on the effect of polydisperse systems in 3D numerical study are necessary. Thirdly, generally negative correlations existed between fouling rate and the hydrodynamics of both phases, but surprisingly the same correlation did not hold between fouling rate and power input.

Keywords: anaerobic fluidized bed membrane bioreactor; granular activated carbon; two-fluid model; membrane fouling mitigation; hydrodynamics

5.1. Introduction

The membrane bioreactor (MBR) is becoming increasingly popular for the treatment of both municipal and industrial wastewater, in part because it requires a smaller equipment footprint to achieve similar or higher permeate purity compared to conventional wastewater treatment process [1]. Unfortunately, the more widespread implementation of MBRs is hampered by extensive fouling, which requires energy-intensive fouling mitigation means [1-3]. To this end, unsteady-state shear means, such as vibration, gas sparging, and particle fluidization, have been assessed as being more energy-efficient [4]. The anaerobic fluidized bed membrane bioreactor (AnFMBR) is a more recent technology that promises high-quality water at a relatively low energy consumption, because of the fluidization of millimeter-sized GAC (granular activated carbon) particles to both absorb the organic foulants and also mechanically scour the membrane surface [5-7].

Previous studies on the AnFMBR have evaluated the impact of pre-treatment process [6, 8-11], temperature [11-13], foulant type [5, 14-16], GAC size [17, 18], or

comparison of GAC and other particle types [19, 20] on the overall performance of the AnFMBR, such as water quality (e.g., chemical oxygen demand (COD), biochemical oxygen demand (BOD), soluble microbial products (SMP)), process behavior (e.g., transmembrane pressure (TMP), permeate flux), and/or costs (e.g., membrane integrity, methane production, total energy consumption). The fluidized GAC particles have been reported to enable lower trans-membrane pressure (TMP) values [5, 6, 19] and fluctuations [19] under both constant-flux and constant-TMP conditions. The mechanical scouring of GAC has been found to reduce TMP by 50 – 70 % compared to fouling mitigation using water shear stress alone during 2.5 days of operation of the AnFMBR [5]. Another study showed that increasing the superficial water velocity from 1 to 2 m/s increased permeate flux and energy consumption by 20 and 58 %, respectively [21]. Yet another study showed that increasing the GAC particle concentration decreased membrane fouling in an AnFMBR up to a concentration of 5 g/L [22]. The relationship between the fluidized GAC particles and fouling extents has been indirectly characterized with an accelerometer, and the optimal GAC size has been reported to be based on a trade-off between particle inertia and energy requirement [23]. In another experimental study, the local hydrodynamics of the fluidized GAC particles (namely, velocity and concentration) with respect to reactor height along the central axis of the membrane surface has been obtained through image analysis [7], which shows the non-uniform distribution of GAC particle velocities and concentrations with height. Because the effectiveness of fouling mitigation is related to the hydrodynamics of both the fluidized GAC particles and water within the reactor [7], it is crucial to understand in detail the associated characteristics throughout the membrane. Although both lab-scale and pilot-scale experiments have ascertained the efficacy of fluidized GAC particles in mitigating fouling, no numerical study has been carried out to date, which can bridge

the gaps in the knowledge base in terms of (i) a comprehensive landscape of the local behaviours, which is labour- and time-intensive experimentally, and (ii) characteristics of the water phase, which is difficult experimentally due to the high GAC particle concentration. This is important for optimizing the mitigation of fouling via GAC fluidization. Computational fluid dynamics (CFD) provides a useful tool for this.

Numerical studies on liquid-solid fluidization have been carried out to understand the impact of numerical parameters (e.g., mesh size and time step [24, 25]), interphase drag correlations [25], liquid properties (e.g., viscosity [24, 26], velocity [24, 26, 27]), and particle properties (e.g., density [24, 26, 28], size [28], polydispersity [28]) on the particle behavior (namely, particle velocity, expanded bed height, bed voidage, segregation, layer inversion, granular temperature) and flow characteristics (namely, fluidization regime, minimum fluidization velocity, transition velocity from homogeneous to heterogeneous fluidization, critical velocity for the layer inversion phenomenon [25]). A study indicated similar expanded bed height and particle concentration between steady-state 2D and 3D simulations of a rectangular liquid-solid fluidized bed, although different dynamic responses during start-up was observed [24]. Regarding axial particle concentration, it has been found to be uniform axially except near the distributor, in the acceleration zone and at the top of expanded bed height [24]. As for radial particle concentration, a uniform profile was found at relatively low liquid velocity [25, 29], while higher liquid velocity caused higher particle concentration at the center column compared to the near-wall region [30]. Both experimental [29] and simulation results [25, 26] showed that particles moved upwards at the center and downwards near the wall, and such radial variations were attenuated for smaller particle sizes and/or density. Largely similar axial particle velocity was found along the reactor height [26]. Generally, particle concentration decreased, while expanded bed height and

axial particle velocity increased with liquid velocity and/or viscosity [25, 26, 30]. Very few studies investigated the correlation between such macro-level parameters and local solid and liquid behavior, despite their important correlation with mechanical scouring efficiency in the AnFMBR [7].

Accordingly, the current study was aimed at bridging the gap by using computational fluid dynamics (CFD) simulations of the AnFMBR to understand the local (i.e., axial and lateral) characteristics of both the fluidized GAC particles and water phases (namely, particle velocity, water velocity, particle concentration) for various particle diameters (namely, in the range of 1.25 – 1.85 mm) and superficial water velocities (namely, ratio of superficial water velocity to minimum fluidization velocity in the range of 6 - 11). The particle momentum and water shear stress were then calculated, after which the relationship with respect to energy requirement was elucidated. The specific objectives were: (i) assess the lateral variations of the GAC particle hydrodynamics; (ii) evaluate the local and overall hydrodynamics of the water phase; (iii) determine the relationship between fouling and the behaviors of both the water and GAC particle phases.

5.2. Experimental and Simulation Methods

The simulation of the fluidization of millimeter-sized GAC particles in water in a two-dimensional (2D) AnFMBR was carried out using the Two-Fluid Model (TFM), which treats both the liquid phase and the GAC particles as continuums, on Multiphase Flow with interphase eXchange (MFiX) 2016-1, an open-source package developed by National Energy Technology Laboratory (NETL) [31]. Fig. 5.1 shows the 2D AnFMBR studied, which had dimensions of 8.9 cm and 19.4 cm in the x - and y -directions, respectively. The dimensions were the same as that used experimentally in Wang et al.

[23] to enable the simulation model to be validated. A 2D study was carried out to focus only on the membrane surface, since the interaction between the membrane surface and the fluidized GAC particles is key to understanding the fouling mitigation efficacy. Furthermore, the shallow depth of AnFMBR column in experiment (i.e, ~2.3 cm) is relatively small compared to column height and width, which indicate 2D simulation is more suitable to mimic such experimental results. The height of the packed bed of GAC particles (h_0) was 5 cm.

Fig. 5.1. Simulation domain of the 2D AnFMBR.

The governing equations used in the current study are listed in Table 5.1, while Table 5.2 and Table 5.3 list the simulation parameters and operating conditions, respectively. Both conservation of mass and conservation of momentum laws are reserved in current study (Table 5.1). The subscripts w and p indicate liquid (water) and solid (monodisperse GAC particle) states, respectively. In the left side of continuity and momentum equations, the first term shows the net rate of accumulation, while the second term indicate the net rate by convection. Current study will focus on hydrodynamics of AnFMBR, as such no mass transfer or chemical reaction is included in the numerical study. Four terms in the right side of momentum equations represent normal and shear force, gravity force, buoyancy force (due to pressure gradient), and

liquid-solid drag force (due to liquid-solid slip velocity), respectively. It is assumed that other liquid-solid interaction forces is negligible, such as added mass force (due to relative acceleration of liquid-solid), Basset force (due to lagging development of boundary layer around solid, relative to solid acceleration), Faxen force (to account the impact of liquid velocity gradient towards added mass force and Basset force) Saffman lift force (due to gradient of liquid velocity), Magnus force (due to solid rotation), and temperature and density gradient forces [31]. Kinetic theory granular flow (KTGF) is used to describe the interaction between particles. According to KTGF, kinetic energy due to particle velocity fluctuation is proportional to the granular temperature of the solid continuum (θ_p). The net rate of granular temperature (see Table 5.1) is affected by diffusive flux of granular temperature ($\overrightarrow{q_{\theta_p}}$), energy dissipation due to inelastic particle collisions (γ_{θ_p}), and transfer of granular temperature between liquid-solid phase (J_{wp}). Further details on the frictional-kinetic solid stress model ($\overline{\overline{S_p}}$) can be found in Srivastava and Sundaresan [32], and on the water stress model ($\overline{\overline{S_w}}$) and fluid-solid momentum transfer (p_{wp}) in Syamlal et al [31]. It should be noted that granular temperature is generally applicable for equilibrium system, while AnFMBR is a transient process which slowly evolve over time. Regardless, Baldassarri et al. [33] has shown that in relation with fluctuation-dissipation theorem and experimental measurement of noise power spectrum using torsion oscillator in a granular bath, the concept of granular temperature can be a priori extended into such slowly evolving system. The impact of turbulence is accounted using *k-epsilon* turbulence model developed by Benyahia et al. [34] with turbulence length scale and maximum turbulent viscosity magnitudes is listed

in Table 5.1. Equations were discretized in a staggered grid [35] and solved with a modified SIMPLE algorithm [36].

Table 5.1. CFD equations used for monodisperse GAC study in 2D AnFMBR.

Continuity Equations	
Continuity equation (liquid)	$\frac{\partial}{\partial t}(\varepsilon\rho_w) + \nabla \cdot (\varepsilon\rho_w \vec{U}_w) = 0$
Solid phase	$\frac{\partial}{\partial t}((1-\varepsilon)\rho_p) + \nabla \cdot ((1-\varepsilon)\rho_p \vec{U}_p) = 0$
Momentum Equations	
Liquid phase	$\frac{\partial}{\partial t}(\varepsilon\rho_w \vec{U}_w) + \nabla \cdot (\varepsilon\rho_w \vec{U}_w \vec{U}_w)$ $= \nabla \cdot \vec{S}_w + \varepsilon\rho_w \vec{g} - \varepsilon \nabla P - I_{wp}$
Solid phase	$\frac{\partial}{\partial t}((1-\varepsilon)\rho_p \vec{U}_p) + \nabla \cdot ((1-\varepsilon)\rho_p \vec{U}_p \vec{U}_p)$ $= \nabla \cdot \vec{S}_p + (1-\varepsilon)\rho_p \vec{g} - (1-\varepsilon) \nabla P + I_{wp}$ <p>where $I_{wp} = \beta(\vec{U}_w - \vec{U}_p)$</p>
Constitutive Equations	
Interphase drag coefficient [37]	$\beta = 150 \frac{(1-\varepsilon)^2 \mu_w}{\varepsilon d_p^2} + 1.75 \frac{(1-\varepsilon)\rho_w \vec{U}_p - \vec{U}_w }{d_p}, \text{ for } \varepsilon < 0.8 \text{ [38]}$ $\beta = \frac{3}{4} C_D \frac{(1-\varepsilon)\rho_w \vec{U}_p - \vec{U}_w }{d_p} \varepsilon^{-1.65}, \text{ for } \varepsilon \geq 0.8 \text{ [39]}$ <p>where $C_D = \frac{24}{Re_p} (1 + 0.15 Re_p^{0.687}), \text{ for } Re_p \leq 1000$</p> $C_D = 0.44, \text{ for } Re_p > 1000$
Granular temperature [40]	$\frac{3}{2} \left[\frac{\partial}{\partial t}((1-\varepsilon)\rho_p \theta_p) + \nabla \cdot ((1-\varepsilon)\rho_p \theta_p \vec{U}_p) \right]$ $= [\vec{S}_p : \nabla \vec{U}_p - \nabla \cdot \vec{q}_{\theta_p} - \gamma_{\theta_p} + I_{wp}]$ <p>where $\theta_p = \frac{1}{3} \langle (\vec{c}_p - \langle \vec{c}_p \rangle)^2 \rangle$</p>

	$\overrightarrow{q_{\theta_p}} = -k_{\theta_p} \nabla \theta_p$ $k_{\theta_p} = \frac{15(1-\varepsilon)d_p\rho_p\sqrt{\pi\theta_p}}{4(41-33\eta)} \left[1 + \frac{12}{5}\eta^2(4\eta-3)(1-\varepsilon)g_0 + \frac{16}{15\pi}(41-33\eta)\eta(1-\varepsilon)g_0 \right]$ $\eta = \frac{1}{2}(1+e_p)$ $\gamma_{\theta_p} = \frac{12(1-e_p^2)\rho_p g_0(1-\varepsilon)^2\theta_p^{3/2}}{d_p\sqrt{\pi}}$
Particle terminal velocity [41]	$U_t = \frac{U_*}{\left[\frac{\rho_w^2}{g\mu_w(\rho_p - \rho_w)} \right]^{1/3}}$ <p>where $U_* = \left(\frac{18}{d_*^2} + \frac{9.3393-6.9756\phi}{4d_*^{0.5}} \right)^{-1}$</p> $d_* = d_p \left[\frac{g\rho_w(\rho_p - \rho_w)}{\mu_w^2} \right]^{1/3}$
Minimum fluidization velocity [39]	$Re_{mf} = (1135.69 + 0.0408Ar)^{1/2} - 33.7$ <p>where $Re_{mf} = \frac{\rho_w U_{mf} d_p}{\mu_w}$ and $Ar = \frac{d_p^3 \rho_w (\rho_p - \rho_w)}{g \mu_w^2}$</p>
Wall boundary condition [42]	$\overrightarrow{U_{p,s}} \cdot \overrightarrow{\bar{S}_p} \cdot \vec{n} + \frac{\sqrt{3}\pi}{6(1-\varepsilon)} \phi'(1-\varepsilon) \rho_p g_{0,pp} \theta_p^{1/2} \overrightarrow{U_{p,s}} ^2 = 0$

Table 5.2. Simulation parameters used for monodisperse GAC study in 2D AnFMBR.

Parameter	Unit	Magnitude
Simulation time	s	200 (only the final 150 s is used for steady-state data analysis)
Time step	s	0.0001
Number of grids ($x \times y$)	-	35×77 (for $d_p = 1.25$ mm) 28×62 (for $d_p = 1.55$ mm) 23×50 (for $d_p = 1.85$ mm)
Water Properties		
Water density (ρ_w)	kg/m ³	1000
Water viscosity (μ_w)	kg/m/s	0.001
Turbulent length scale (l)	m	0.00336
Maximum turbulent viscosity (μ_{wt})	kg/m/s	100
GAC Properties		
GAC particle density (ρ_p)	kg/m ³	1500
GAC particle diameter (d_p)	mm	1.25, 1.55, 1.85
Particle-particle restitution coefficient (e_p)	-	0.99
Particle-wall restitution coefficient (e_r)	-	0.99
Angle of internal friction (ϕ)	°	30
Angle of internal friction at wall (ϕ_r)	°	11.31
Specularity coefficient at wall (ϕ') [43]	-	0.0001
Initial Conditions		
Particle Bed		

Initial bed height (h_0)	m	0.05
Initial void fraction (ε_b)	-	0.5
Freeboard		
Void fraction (ε_f)	-	1 (i.e., no GAC particles)
Boundary Conditions		
Bottom ($y/Y = 0$): uniform superficial liquid velocity (U_l)	m/s	0.0258 – 0.0827
Top ($y/Y = 1$): uniform water pressure	Pa	101325
Sides ($x/X = 0$ and 1): partial slip for particle-wall interaction	-	Johnson-Jackson boundary conditions [42]

Table 5.3. Operating conditions used for monodisperse GAC study in 2D AnFMBR.

d_p (mm)	U_{mf} [39] (m/s)	U_t [41] (m/s)	U_l (m/s)			
			$\frac{U_l}{U_{mf}} = 6$	$\frac{U_l}{U_{mf}} = 7$	$\frac{U_l}{U_{mf}} = 9$	$\frac{U_l}{U_{mf}} = 11$
1.25	0.00429	0.0696	0.0258	0.0301	0.0386	0.0472
1.55	0.00623	0.0812	0.0374	0.0436	0.0561	0.0686
1.85	0.00827	0.0914	0.0496	0.0579	0.0745	-*

*Not investigated due to particle entrainment at this condition

Fig. 5.2 shows the contour plots of bed void fraction (ε) during the fluidization of GAC particles with $d_p = 1.25$ mm at a ratio of superficial liquid velocity to minimum fluidization velocity (U_l/U_{mf}) of 6 at time (t) between 0 – 200 s. Initially between $t = 0$ – 0.1 s (Figs. 5.2a and b), the GAC particles were in a packed bed with uniform bed void fraction (ε) throughout the bed. At around $t = 1$ s (Fig. 5.2c), the lower bed void fraction

(ϵ) values were at the top and sides. Subsequently at $t = 5 - 10$ s (Figs. 5.2d - e), the higher bed void fraction (ϵ) values took the form of an hour-glass shape, which indicates liquid channeling behavior mid-height of the expanded particle bed. Finally, at $t \geq 50$ s (Figs. 5.2f - h), the hour-glass shape of higher bed void fraction (ϵ) values became constant with time. Collectively, the contour plots in Fig. 5.2 indicates that the void fraction (ϵ) evolved with time before attaining steady-state at approximately 50 s (Fig. 5.2f), hence data-averaging of the variables of interest was only carried out for the final 150 s (i.e., $t = 51 - 200$ s). Notably, the liquid channeling behavior is disadvantageous, since particles are key to affecting the mechanical scouring for fouling mitigation. Although uniform bed void fraction tends to be more prevalent in liquid-solid fluidization than gas-solid fluidization, the non-uniformity per Fig. 5.2 is also well acknowledged [44-46]. Fig. 5.2. is not designed to evaluate detailed local particle concentration, as such mean is not the main concern of membrane community on AnFMBR system (i.e., the focus is overall fouling effectiveness evaluation and energy efficiency compared to the more common gas sparging system).

Fig. 5.2. Contour plots of bed void fraction (ϵ) for GAC particles with $d_p = 1.25$ mm and $U/U_{mf} = 6$ at $t =$ (a) 0 s, (b) 0.1 s, (c) 1 s, (d) 5 s, (e) 10 s, (f) 50 s, (g) 100 s, and (h) 200 s.

To ensure the validity of the parameters used in the TFM simulation, it is necessary to ensure the simulation results agree with experimental data. Wang et al. [23] has earlier reported on high-speed video camera images focused on the plane of the membrane surface to understand the particle hydrodynamics (namely, particle velocity, concentration, and momentum) in relation to the extent of fouling mitigation. This experimental dataset was used to validate the TFM model in this study. Fig. 5.3 compares the local average of particle velocity ($U_{p, ave}$) and particle volume fraction ($1 - \varepsilon$) at various vertical positions (y/Y , where y is the height assessed and Y is the total height of the setup; y/Y values of 0 and 1 refer respectively to the bottom and top edges of the AnFMBR) along $x/X = 0.5$ (where x is the lateral position assessed and X is the total width of the setup; x/X values of 0 and 1 refer respectively to the left and right edges of the AnFMBR) between the experimental [23] and simulated results. Figs. 5.3a and b contain the $U_{p, ave}$ trends with respect to U_l/U_{mf} for GAC particle diameters of 1.55 mm and 1.85 mm, respectively. Model validation was limited to the higher heights of $y/Y \geq 0.57$ of the reactor to avoid the inevitable entrance effects in the experiments, which depends on the specific design of the liquid distributor and causes the hydrodynamics lower in the column to vary among setups [47, 48]. Since the current TFM model assumed an idealized uniform inlet flow (Table 5.2), the comparison with the experimental data was only carried out after mid-height of the reactor to evade the entrance effects in the experimental setup. Fig. 5.3 shows that the simulation results are of the same order-of-magnitude and have the same trends with respect to U_l/U_{mf} as the experimental data. The averagely-lower $U_{p, ave}$ in simulation results compared to experimental data can be attributed to the reduced reactor depth in middle-upper section of experiment set up due to the presence of membrane module [23]. Specifically, reactor depth is 3.3 cm and 1.7 cm for reactor height 0 – 4.4 cm and 4.4 – 19.4 cm, respectively.

Reduced reactor depth increases lateral average water velocity at middle-upper column compared to inlet superficial water velocity measured at liquid distributor. Regrettably, the impact of reactor depth cannot be measured in current 2D numerical study. Fig. 5.3 also indicates the good agreement between expansion bed height in experimental [23] and simulation studies. Both experimental and simulation studies found the relatively low bed expansion height ($y/Y < 0.57$, indicated by 0 particle velocity) for 1.55 and 1.85 mm GAC particles at U_l/U_{mf} of 7.60 and 5.72 – 6.44, respectively.

Fig. 5.3. Comparison between simulation and experimental [23] results at $x/X = 0.5$ of average particle velocity ($U_{p,ave}$) with GAC particle diameters (d_p) of (a) 1.55 mm and (b) 1.85 mm.

5.3. Results and Discussion

5.3.1. Local particle velocity

Figs. 5.4 – 5.6 assess the effect of the ratio of superficial water velocity to the minimum fluidization velocity (U_l/U_{mf}) and GAC particle diameter (d_p) on local particle velocities (U_p) at various heights (y/Y) along the reactor. Specifically, Figs. 5.4 – 5.6 present respectively the local y -directional particle velocity ($U_{p,y}$), local x -directional

particle velocity ($U_{p,x}$), and local average of particle velocity ($U_{p,ave}$) with respect to x/X (i.e., the reactor width). Each figure contains 9 sub-plots, with each of the three columns representing each of the three GAC particle diameters (namely, $d_p = 1.25$ mm, 1.55 mm, and 1.85 mm), and each of the three rows representing each height along the reactor (namely, $y/Y = 0.3, 0.5$, and 0.8). Each of the 9 sub-plots displays four trends for the two smaller particles ($d_p = 1.25$ and 1.55 mm) and three trends for the largest particles ($d_p = 1.85$), which represent velocity ratios of U/U_{mf} in the range of 6 – 11. The rightmost column of sub-plots for the largest particles ($d_p = 1.85$ mm) only contained three trends because the highest U/U_{mf} of 11 was not possible due to entrainment. The axes ranges in each figure were largely kept the same for an easier comparison. It should be noted that the particle velocity profiles are that on the plane of the membrane surface, hence indicates the extent of the particle – membrane surface interactions.

Fig. 5.4 describes the local y -directional particle velocity ($U_{p,y}$) with respect to the reactor width (x/X) for various GAC particle diameters (d_p), reactor heights (y/Y), and dimensionless superficial water velocities (U/U_{mf}). Positive and negative values of $U_{p,y}$ indicate respectively up-flowing and down-flowing particles. Regarding the mechanical scouring behavior of particles leading to membrane fouling mitigation, the magnitude rather than the direction (upward or downward) is expected to determine the extent of fouling mitigation.

Fig. 5.4. Local y -directional particle velocity ($U_{p,y}$) with respect to reactor width (x/X) at various superficial water velocities ($U_l/U_{mf} = 6 - 11$) and axial positions ($y/Y = 0.3 - 0.8$) for GAC particle diameters (d_p) of (a) 1.25 mm, (b) 1.55 mm, and (c) 1.85 mm.

Four observations regarding the $U_{p,y}$ profiles (Fig. 5.4) are worth noting here. Firstly, the profiles were either flat at $U_{p,y} = 0$ (in the absence of fluidized particles), or inverted V-shaped (only for the smallest particles of $d_p = 1.25$ mm at the lowest height investigated of $y/Y = 0.3$), or the most prevalent M-shaped, which agrees with previous experimental studies [44, 49, 50]. Both the inverted V-shaped and M-shaped profiles indicate that fouling mitigation was non-uniform laterally across the membrane, particularly with some lateral positions lacking any mitigation effects where $U_{p,y} = 0$.

Secondly, the maximum magnitudes of $U_{p,y}$, and thereby the greatest extent of y -directional fouling mitigation, were found near the walls ($x/X \approx 0.05$ and 0.95) for the two lower positions ($y/Y = 0.3$ and 0.5), whereas relatively more uniformly distributed laterally at the highest height investigated ($y/Y = 0.8$). Thirdly, the increase of U/U_{mf} expectedly resulted in larger local and overall magnitudes of $U_{p,y}$, which thereby affirmed the positive relationship between energy input (correlated to U/U_{mf}) and fouling mitigation effectiveness [7, 23]. Fourthly, on the impact of d_p , the profiles of the two larger particles ($d_p = 1.55$ and 1.85 mm) appear similar, whereas that of the smallest particle ($d_p = 1.25$ mm) appear quite different. This agrees with a previous study, which reported that the correlation between scouring efficiency and particle diameter (d_p) was positive at the lower d_p range but neutral at the upper d_p range [7]. Collectively, considering the significant variations which would lead to non-uniform fouling of the membrane, the observations underscore the importance of understanding the local behavioral characteristics during the process optimization of the fluidized bed – membrane reactor.

Analogous to Fig. 5.4, Fig. 5.5 presents the trends for the x -directional velocities ($U_{p,x}$) with respect to reactor width (x/X). The x -directional particle velocities ($U_{p,x}$) in Fig. 5.5 had considerably smaller magnitudes compared to $U_{p,y}$ (Fig. 5.4), with the exception of the smallest particles ($d_p = 1.25$ mm) at the lowest height investigated ($y/Y = 0.3$), which corresponds to the only inverted V-shaped $U_{p,y}$ profiles (Fig. 5.4a). More explicitly, the high magnitudes of $U_{p,y}$ (Fig. 5.4a) at the reactor center ($x/X = 0$) for the smallest particles ($d_p = 1.25$ mm) at the lowest height ($y/Y = 0.3$) caused a strong lateral dispersion of the peripheral particles towards the wall, which is reflected in the high magnitudes of $U_{p,x}$ at approximately $x/X = 0.25$ and 0.75 (Fig. 5.5a). For the smaller particles (Figs. 5.5a and b), where slight magnitudes of $U_{p,x}$ values appear, it is correlated

with flatter $U_{p,y}$ profiles (i.e., $U_{p,y} \approx 0$ laterally; Fig. 5.4), which correspond to the top of the expanded bed height and represent the height at which the upward inertial force supplied by the liquid on the particles was depleted. In particular for the largest particles ($d_p = 1.85$ mm), the $U_{p,x}$ values were negligible throughout the reactor due to the greater upwards (y) inertia, which indicate negligible lateral scouring.

Fig. 5.5. Local x -directional particle velocity ($U_{p,x}$) with respect to reactor width (x/X) at various superficial water velocities ($U_l/U_{mf} = 6 - 11$) and axial positions ($y/Y = 0.3 - 0.8$) for GAC particle diameters (d_p) of (a) 1.25 mm, (b) 1.55 mm, and (c) 1.85 mm.

In addition to the x - ($U_{p,x}$; Fig. 5.3) and y -directional ($U_{p,y}$; Fig. 5.4) velocities, Fig. 5.6 describes the average particle velocity ($U_{p,ave}$), which can be calculated by:

$$U_{p,ave} = \sqrt{U_{p,x}^2 + U_{p,y}^2} \quad (1)$$

In contrast to Figs. 5.4 and 5.5, the $U_{p,ave}$ values summarizes the velocities in all directions to reflect the scouring capacity induced by particle velocity (U_p). Four observations are worth highlighting. Firstly, $U_{p,ave}$ (Fig. 5.6) magnitudes were similar to that of $U_{p,y}$ (Fig. 5.4), which affirms the dominance of the y -directional velocities in inducing the mechanical scouring effects. Secondly, the profiles were largely of the form of two asymmetrical W-shaped profiles side-by-side, which indicate that the lateral distribution of $U_{p,ave}$ values was uneven. This indicates the non-uniform fouling mitigation effectiveness induced by particle velocity (U_p) across the membrane surface and thereby emphasizes the necessity of knowing the local information for enhanced predictive capability of the membrane process performance. Thirdly, $U_{p,ave}$ generally increased with water velocity (U_l/U_{mf}), with the exception again of the smallest particle ($d_p = 1.25$ mm) at the lowest height ($y/Y = 0.3$) investigated, whereby the lower U_l/U_{mf} values of 6–7 surprisingly gave higher $U_{p,ave}$ values. The exception is due to the particle concentration variation along the membrane height, which is explored in Fig. 5.7. For the case of $U_l/U_{mf} = 6–7$ for the smallest particles ($d_p = 1.25$ mm), the particles were only at the lower heights and completely absent from the higher heights ($y/Y \geq 0.5$), thereby all the energy transferred from the up-flowing water to the particles was concentrated at the lower heights, which led to higher $U_{p,ave}$ values. This agrees with the observations in a previous study [23]. Fourthly, at the similar U_l and thereby energy requirement for the different particle diameters (d_p), the larger particles gave slightly lower $U_{p,ave}$ values, which agree with a previous study [7]. For examples, the $U_{p,ave}$

values for $d_p = 1.25$ mm and $U_l/U_{mf} = 9$ were larger than those of $d_p = 1.55$ mm and $U_l/U_{mf} = 6$, and the $U_{p,ave}$ values of $d_p = 1.55$ mm and $U_l/U_{mf} = 9$ were larger than those of $d_p = 1.85$ mm and $U_l/U_{mf} = 7$. This phenomenon is presumably due to the trade-off between particle inertia and $U_l - U_{mf}$, specifically in that, at the same U_l , the $U_l - U_{mf}$ value and thereby the mobility of the particles is greater for the smaller particles.

Fig. 5.6. Local average particle velocity ($U_{p,ave}$) with respect to reactor width (x/X) at various superficial water velocities ($U_l/U_{mf} = 6 - 11$) and axial positions ($y/Y = 0.3 - 0.8$) for GAC particle diameters (d_p) of (a) 1.25 mm, (b) 1.55 mm, and (c) 1.85 mm.

5.3.2. Particle volume fraction

In addition to only the concentration variations along the height experimentally investigated in the previous study [7], the current study assessed the lateral variations. Both particle concentrations and particle velocities are important in mitigating fouling, since high magnitudes of particle velocities (U_p) are inadequate in effective scouring if the particle concentration is low. Fig. 5.7 displays the local particle volume fraction as $(1 - \varepsilon)$, where ε represents void fraction. Three key observations from Fig. 5.7 are discussed here. Firstly, regardless of height along the membrane (y/Y) or particle diameter (d_p) or superficial water velocity (U_l/U_{mf}), the particle volume fraction ($1 - \varepsilon$) was approximately constant along a large part of the reactor laterally ($0.2 \leq x/X \leq 0.8$) but decreased rapidly near the walls. This indicates that the particle concentrations near the wall were insignificant, therefore, even though the particle velocities were high near the wall (Figs. 5.4 and 5.6), fouling mitigation is expected to be limited. Secondly, particle concentration ($1 - \varepsilon$) generally decreased with U_l/U_{mf} due to progressively greater bed expansion. The only exception to this was for $d_p = 1.55$ mm at $y/Y = 0.5$ (Fig. 5.7b), whereby the particle concentration values for $U_l/U_{mf} = 6$ were lower than those for $U_l/U_{mf} = 7$. This is presumably because $y/Y = 0.5$ was approximately the expanded bed height for the former condition, thereby reflected the lowered concentration of both the dense bed and the dilute region above the bed. Thirdly, within the fluidized bed, the particle concentration only decreased slightly with height, indicating an approximately uniform distribution of particles throughout the fluidized bed except near the walls.

Fig. 5.7. Local particle volume fraction ($1 - \epsilon$) with respect to reactor width (x/X) at various superficial water velocities ($U_l/U_{mf} = 6 - 11$) and axial positions ($y/Y = 0.3 - 0.8$) for GAC particle diameters (d_p) of (a) 1.25 mm, (b) 1.55 mm, and (c) 1.85 mm.

5.3.3. Local water velocity

Liquid shear on the membrane surface is well acknowledged to mitigate fouling. Other than the mechanical scouring effects by the fluidized GAC particles, the water phase also provides tangential shear forces on the membrane surface, both of which serve to mitigate membrane fouling concurrently. This section investigates the local

water velocity for the various particle diameters (d_p), superficial water velocities (U_l) and thereby energy requirement, and positions along the membrane (x/X and y/Y).

Fig. 5.8 shows the local y -directional water velocity ($U_{w,y}$) with respect to x/X . Five key observations are noted as follows. Firstly, as opposed to the y -directional particle velocity ($U_{p,y}$), the $U_{w,y}$ values were all positive, which implied only upward motion. Secondly, the higher U_l/U_{mf} values gave M-shaped profiles while the lower U_l/U_{mf} values gave inverted-U or inverted-V profiles. While the M-shaped profiles gave the highest $U_{w,y}$ values in between the center and walls ($x/X \approx 0.2$ and 0.8), the inverted-U or V profiles gave the highest $U_{w,y}$ values around the center ($x/X \approx 0.5$). Similar to that for the particles, the liquid velocities were also non-uniform laterally across the membrane. Thirdly, the peak $U_{w,y}$ magnitudes were greater than the corresponding U_l values employed, presumably because the presence of GAC decreased the cross-sectional area for y -directional water flow. Fourthly, the lowest $U_{w,y}$ values tended to be near the walls, presumably due to wall friction. Fifthly, the shapes of the $U_{w,y}$ and $U_{p,y}$ profiles are very similar, which indicate similar y -directional behaviors of the two phases.

Fig. 5.8. Local y -directional water velocity ($U_{w,y}$) with respect to reactor width (x/X) at various superficial water velocities ($U_l/U_{mf} = 6 - 11$) and axial positions ($y/Y = 0.3 - 0.8$) for GAC particle diameters (d_p) of (a) 1.25 mm, (b) 1.55 mm, and (c) 1.85 mm.

Fig. 5.9 shows the local x -directional water velocity ($U_{w,x}$) with respect to x/X . The positive and negative values of $U_{w,x}$ indicate respectively rightward and leftward directions of the local water motion. Three interesting observations are worth highlighting. Firstly, the maximum $U_{w,x}$ values were one order-of-magnitude lesser than the $U_{w,y}$ values (Fig. 5.8), which is not surprising since water is directed upwards from the bottom. This is similar to that for $U_{p,x}$, and implies that the shear in the lateral

direction is much weaker than that in the vertical direction. Secondly, the greatest $U_{w,x}$ values are found at the lowest y/Y of 0.3 for the smallest d_p of 1.25 mm, which agree with that for $U_{p,x}$. Thirdly, the $U_{w,x}$ and $U_{p,x}$ profiles are similar, which indicate the similar behaviors of the two phases. This agrees with that for the y -direction.

Fig. 5.9. Local x -directional water velocity ($U_{w,x}$) with respect to reactor width (x/X) at various superficial water velocities ($U_l/U_{mf} = 6 - 11$) and axial positions ($y/Y = 0.3 - 0.8$) for GAC particle diameters (d_p) of (a) 1.25 mm, (b) 1.55 mm, and (c) 1.85 mm.

Fig. 5.10 evaluates the local average water velocity ($U_{w,ave}$) along the reactor width (x/X) at various axial positions (y/Y) and superficial water velocities (U_l/U_{mf}).

$U_{w,ave}$ was obtained by:

$$U_{w,ave} = \sqrt{U_{w,x}^2 + U_{w,y}^2} \quad (2)$$

Five observations are noted. Firstly, unsurprisingly, the $U_{w,ave}$ magnitudes and trends in Fig. 5.10 mimic those of $U_{w,y}$ (Fig. 5.8) rather than $U_{w,x}$ (Fig. 5.9), because of the order-of-magnitude difference in magnitudes between $U_{w,y}$ and $U_{w,x}$. Secondly, although the shapes of the profiles are similar between $U_{p,y}$ (Fig. 5.4) and $U_{w,y}$ (Fig. 5.8), and also $U_{p,x}$ (Fig. 5.5) and $U_{w,x}$ (Fig. 5.9), the profiles of $U_{p,ave}$ (Fig. 5.6) and $U_{w,ave}$ (Fig. 5.10) are distinctly different. This is because the $U_{w,y}$ (Fig. 5.8) values were all positive, whereas $U_{p,y}$ (Fig. 5.4) had both positive and negative values. Thirdly, among the results at similar water velocity (U_l) and thereby similar energy requirement (namely, $U_l/U_{mf} = 11$ for $d_p = 1.25$ mm, $U_l/U_{mf} = 7$ for $d_p = 1.55$ mm, and $U_l/U_{mf} = 6$ for $d_p = 1.85$ mm (Table 5.3), the $U_{w,ave}$ trends and magnitudes were very similar at all y/Y positions, which imply the negligible impact of d_p . Fourthly, increasing the energy requirement through increasing U_l was ineffective in enhancing the water shear on the membrane near the walls (i.e., $x/X = 0$ and 1) and center (i.e., $x/X = 0.5$). Fifthly, the shapes of the $U_{w,ave}$ profiles remain approximately constant for $U_l/U_{mf} \geq 9$, except at the highest y/Y position (i.e., $y/Y = 0.8$) for $d_p = 1.25$ mm.

Fig. 5.10. Local average water velocity ($U_{w,ave}$) with respect to reactor width (x/X) at various superficial water velocities ($U_l/U_{mf} = 6 - 11$) and axial positions ($y/Y = 0.3 - 0.8$) for GAC particle diameters (d_p) of (a) 1.25 mm, (b) 1.55 mm, and (c) 1.85 mm.

5.3.4. Particle momentum

To relate the behavior of the particles with fouling mitigation effectiveness, it has been reported that particle momentum (p), which accounts for both particle velocity ($U_{p,ave}$) and particle concentration ($1 - \varepsilon$), is a better indicator than $U_{p,ave}$ or $(1 - \varepsilon)$ alone [23]. This is because both play important roles in effecting the mechanical scouring of the membrane. Specifically, particle momentum (p) can be quantified by:

$$p = U_{p,ave} \cdot \rho_p \cdot (1 - \varepsilon) \cdot \Delta V \quad (3)$$

where ρ_p is particle density and ΔV is the volume of the portion of the reactor assessed. Accordingly, Fig. 5.11 quantifies the GAC particle momentum (p) with respect to reactor width (x/X) at various axial positions (y/Y) and superficial water velocities (U/U_{mf}). Three key observations from Fig. 5.11 are as follows. Firstly, for all the conditions assessed, the greatest particle momentum (p) and thereby most effective membrane fouling mitigation was for the smallest particle diameter ($d_p = 1.25$ mm), lowest height ($y/Y = 0.3$) and the lower superficial water velocities ($U/U_{mf} = 6$ and 7). This is due to the coupling of both high $U_{p,y}$ (Fig. 5.4a) and high $1 - \varepsilon$ (Fig. 5.7a) values. Secondly, the most common profile shape is that with three distinct peaks, and exist consistently for all particle diameters (d_p) and heights (y/Y) when $U/U_{mf} \geq 9$. Such profiles indicate very non-uniform lateral fouling mitigation extents and the most effective fouling mitigation was at approximately $x/X \approx 0.2, 0.5$ and 0.8 . Thirdly, the effect of U/U_{mf} on particle momentum (p) is not clear, due to the opposite effects of U/U_{mf} on particle velocity ($U_{p,ave}$) and particle volume fraction ($1 - \varepsilon$).

Fig. 5.11. Local particle momentum transfer (p) with respect to reactor width (x/X) at various superficial water velocities ($U_l/U_{mf} = 6 - 11$) and axial positions ($y/Y = 0.3 - 0.8$) for GAC particle diameters (d_p) of (a) 1.25 mm, (b) 1.55 mm, and (c) 1.85 mm.

5.3.5. Water shear stress

The efficacy of the commonly implemented cross-flow in fouling mitigation is rooted in the shear stress (τ_w) induced on the membrane surface. Shear stress (τ_w) can be derived by:

$$\tau_w = \mu \frac{\partial U_{w,ave}}{\partial y} \quad (4)$$

where $\frac{\partial U_{w,ave}}{\partial y}$ is the change in $U_{w,ave}$ with respect to the axial positions (y); ∂y indicates individual grid height, i.e., 2.52 mm, 3.12 mm, and 3.88 mm for the GAC particles with particle diameters (d_p) of 1.25 mm, 1.55 mm, and 1.85 mm, respectively. Fig. 5.12 presents the local water shear stress (τ_w) trends and thereby the extent of fouling mitigation conferred by the water phase. Four insights are obtained. Firstly, the maximum water shear stress (τ_w) values were predominantly near the wall ($x/X \approx 0$ and 1), whereas the minimum values were around the center ($x/X \approx 0.5$). This contrasts with the particle momentum trends, whereby the maximum and minimum p values were instead near the center and wall, respectively. Secondly, the τ_w profiles are largely invariant with height along the reactor (y/Y). Thirdly, the τ_w values generally increased monotonically with U_l/U_{mf} , with the effect being more pronounced as the particle diameter (d_p) increased. Fourthly, at the same U_l and thereby energy requirement (namely, comparing the two conditions of $d_p = 1.25$ mm and $U_l/U_{mf} = 9$, and $d_p = 1.55$ mm and $U_l/U_{mf} = 6$; and also the two conditions of $d_p = 1.55$ mm and $U_l/U_{mf} = 9$, and $d_p = 1.85$ mm and $U_l/U_{mf} = 7$), the larger particles tended to exhibit slightly lower τ_w values. This contradicts that of the local water velocity ($U_{w,ave}$), which is due to the smaller lateral variation of $U_{w,ave}$ for the larger particles (Fig. 5.10).

Fig. 5.12. Local water shear stress (τ_w) with respect to reactor width (x/X) at various superficial water velocities ($U_l/U_{mf} = 6 - 11$) and axial positions ($y/Y = 0.3 - 0.8$) for GAC particle diameters (d_p) of (a) 1.25 mm, (b) 1.55 mm, and (c) 1.85 mm.

5.3.6. Energy requirement

The optimization of the fluidized bed – membrane reactor depends on the trade-off between energy requirement and fouling mitigation effectiveness [4]. While higher energy input typically gives higher fouling mitigation extents, the energy costs also escalates, thus optimization is necessary. The power requirement is expressed as:

$$\text{Power} = U_l \cdot A \cdot \Delta P \quad (5)$$

where U_l is the superficial liquid velocity, A is the cross-sectional area of the reactor, and ΔP is the pressure drop across the expanded particle bed. Accordingly, Figs. 5.13a and b plot the arithmetic averages of respectively the water shear stress (τ_w) and particle momentum (p) values laterally (x) versus power requirement (equation $\text{Power} = U_l \cdot A \cdot \Delta P$ (5)). Specifically, water shear stress (τ_w) and particle momentum (p) represent the contributions of respectively the water and GAC particle phases in effecting the membrane fouling mitigation. Although it is in principle possible to designate a value for the particle shear stress (τ_p), it is not straightforward and thus not attempted in this study for two chief reasons [51]: (i) it would include the water phase and also requires an assumption of the viscosity of the fluidized GAC suspension; and (ii) the suspension exhibits non-Newtonian behavior, which implies that the apparent viscosity is not a constant but rather a function of shear rate. Fig. 5.13 reveals that (i) expectedly both τ_w and p increased with power; (ii) higher τ_w and p magnitudes were possible for the larger particles, but at the cost of higher power; (iii) to attain the same τ_w or p values, the power required was greater for the larger particles, which indicate that the smaller particles are more energy-efficient; and (iv) at the same power input, the τ_w or p values for the different particle diameters (d_p) overlapped, which indicates that adequate control of parameters other than particle diameter (d_p) can confer similar shearing or scouring effects. Fig. 5.14 further plots water shear stress (τ_w) versus particle momentum (p) for different GAC particle diameters (d_p). Although a positive correlation is clear between τ_w and p for each particle diameter (d_p), the spread of p values at each τ_w or the spread of τ_w values at each p tended to be significant when all three particles are considered collectively. Notably, for the same water shear stress (τ_w), the particle momentum (p) was consistently greater for the larger particles. Furthermore, Figs. 5.15a – d show the relation between fouling rate ($dTMP/dt$) obtained from Wang et al. experimental data

[23] versus current simulation results on lateral average of local average particle velocity ($U_{p, ave}$), particle concentration ($1 - \epsilon$), particle momentum (p), and power input. Three finding worth mentioning here. First, simulation results on lateral average local data (i.e., $U_{p, ave}$, $1 - \epsilon$, p , and power input) and fouling rate data of experimental results has clear trend, compared to local data at centre column versus fouling rate (see Wang et al. [23]), which implies the importance of complete local data on hydrodynamics evaluation of AnFMBR. Second, fouling rate decreases as $U_{p, ave}$, $1 - \epsilon$, p , or power input increases. Third, larger particles may decrease fouling rate at the cost of higher power input.

Fig. 5.13. (a) Water shear stress (τ_w) and (b) particle momentum transfer (p) as function of power input.

Fig. 5.14. Particle momentum (p) versus water shear stress (τ_w).

Furthermore, the relationship between the simulation results obtained here and the fouling characteristics was sought. In Fig. 5.15, the y-axes represent the fouling rates in terms of the rate of change of transmembrane pressure ($dTMP/dt$) obtained by Wang et al. [23] experimentally at various heights. The x-axis of each of the four sub-plots in Fig. 5.15 represent laterally averaged local values obtained in the current simulation, namely, the average particle velocity ($U_{p,ave}$), particle concentration ($1 - \epsilon$), particle momentum (p), average water velocity ($U_{w,ave}$), water shear stress (τ_w) and power input. Three interesting findings are worth noting. Firstly, in contrast to the experimental results whereby the correlations between $dTMP/dt$ and either $U_{p,ave}$, $(1 - \epsilon)$ and p were weak [23], the additional lateral information obtained in the current simulation study served to show the expected negative correlation between $dTMP/dt$ and all of $U_{p,ave}$, $(1 - \epsilon)$ and p . This is presumably because the hydrodynamics was only characterized at one lateral position per height experimentally [23], which is deficient in accounting for the distinctly non-uniform lateral trends shown in the current study. In view of the significant variations in the hydrodynamics both axially and laterally, a comprehensive landscape of the local information, as obtained here, is crucial for providing the necessary operational and design heuristics. Secondly, negative correlations were also obtained between $dTMP/dt$ and both $U_{w,ave}$ and τ_w . It should be noted that the information on the water phase has not been reported to date. Thirdly, in contrast to the experimental results which showed a general negative correlation between fouling rate and power [23], the relationship was very weak according to the simulation results here. This implies that increasing the power input is ineffective in mitigating fouling.

Fig. 5.15. Fouling rate ($dTMP/dt$) [23] versus (a) local average particle velocity ($U_{p,ave}$), (b) particle concentration ($1 - \epsilon$), (c) particle momentum (p), and (d) power input.

5.4. Conclusions

The fluidization of GAC particles in an AnFMBR has been acknowledged as an effective means for mitigating membrane fouling. A couple of studies have sought to experimentally understand the relationship between the hydrodynamics of the fluidized

GAC particles and the extent of fouling mitigation [7, 23]. However, experimental efforts to provide a comprehensive landscape of the particle hydrodynamics (i.e., particle (U_p) and water (U_l) velocities in the vertical (y) and lateral (x) directions, particle concentration ($1 - \varepsilon$)) via for example the high-speed video camera technique [23] would not only be labor- and time-intensive, but information on the water phase via techniques like particle image velocimetry (PIV) is also encumbered by the high concentrations of GAC. CFD (computational fluid dynamics) provides a useful tool for obtaining a complete landscape of the local particle and water hydrodynamics, in order to better optimize and design the AnFMBR.

The two-fluid model (TFM) used here was first validated against previous experimental data [23], and found to agree well. Local characteristics of the GAC particles and water phases were then obtained throughout the reactor. Subsequently, the more practical performance indicators of particle momentum, water shear stress and power requirement were correlated. Three conclusions are worth highlighting. Firstly, the larger particles are more effective for fouling mitigation, i.e., provide greater particle momentum at similar water shear stress magnitude, at the expense of higher power of pump requirement. On the other hand, smaller particles induce smaller particle momentum while they can be dispersed at lower superficial water velocity (i.e., lower power input). Secondly, for similar power input, different particle diameters may provide similar water shear stress or particle momentum, which indicate other parameter (e.g., superficial water velocity) can be manipulated to improve AnFMBR performance using fixed particle diameter. Thirdly, the correlations between the fouling rate ($dTMP/dt$) and each characteristic – namely, average particle velocity ($U_{p,ave}$), particle concentration ($1 - \varepsilon$), particle momentum (p), average water velocity ($U_{w,ave}$), water shear stress (τ_w) – were generally negative. However, the same correlation did not hold

between fouling rate ($dTMP/dt$) and power input, which indicates that increasing power is ineffective in increasing the mitigation of membrane fouling.

It should be mentioned that the monodisperse (i.e., single particle sizes) particles studied here represents ideal systems, which provides a mechanistic understanding of the behavior of the system, but future studies on the inevitably polydisperse (i.e., range of particle sizes) particles in practical systems will be useful. Furthermore, future work will employ 3D numerical study to provide more-quantitative-based results.

Nomenclature

A	= reactor cross-section area, m^2
Ar	= Archimedes number, –
$\vec{c}_p$	= instantaneous translational particle velocity, m/s
d^*	= dimensionless particle diameter, –
d_p	= particle diameter, mm
e_p	= particle's coefficient of restitution, –
g	= gravitational acceleration, m/s^2
g_0	= equilibrium radial distribution function at contact, –
J_{wp}	= granular temperature transfer between water and solid, $kg/m/s^3$
L	= turbulent length scale, m
m_s	= mass of total particles, kg
$\vec{n}$	= unit normal vector from a boundary to particle, –
p	= momentum transfer of particle, $kg.m/s$
$q_{\theta p}$	= diffusive flux of granular temperature, kg/s^3
Re_{mf}	= Reynolds number at minimum fluidization, –
Re_w	= Reynolds number of water, –
U_*	= dimensionless particle velocity, –
U_l	= inlet superficial water velocity, m/s
U_{mf}	= minimum fluidization velocity, m/s
$U_{p,ave}$	= local average of particle velocity, m/s
$U_{p,s}$	= slip particle velocity = $U_w - U_p$, m/s
$U_{p,x}$	= local x-directional particle velocity, m/s
$U_{p,y}$	= local y-directional particle velocity, m/s
U_t	= terminal velocity of particle, m/s
$U_{w,ave}$	= local average of water velocity, m/s
$U_{w,x}$	= local x-directional water velocity, m/s
$U_{w,y}$	= local y-directional water velocity, m/s
x	= lateral position in reactor, m
X	= reactor width, m
y	= axial position in reactor, m

Y = reactor height, m

Symbol and Greek Letters

ΔP = pressure drop across fluidized bed, Pa

ΔV = differential volume assessed, m³

ε = reactor void fraction, –

ε_b = initial packed bed void fraction ($t = 0$ s), –

ε_f = initial freeboard void fraction ($t = 0$ s), –

μ_w = water viscosity, kg/m/s

μ_{wt} = maximum turbulent viscosity, kg/m/s

θ_p = granular temperature, m²/s²

$\emptyset$ = angle of internal friction, °

$\emptyset'$ = specularity coefficient at wall, –

$\emptyset_r$ = angle of internal friction at wall, °

ρ_w = water density, kg/m³

ρ_p = particle density, kg/m³

γ_{θ_p} = rate of dissipation of granular energy, kg/m/s³

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CHAPTER 6: THE IMPACT OF BIDISPERSE GAC FLUIDIZATION ON MEMBRANE FOULING MITIGATION IN ANFMBR

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Abstract

The anaerobic fluidized-bed membrane bio-reactor (AnFMBR) makes use of fluidized granular activated carbon (GAC) to mechanically scour the membrane to mitigate fouling. The question addressed in the current study is whether fluidizing bidisperse mixtures (i.e., particles of two different sizes) can improve the energy-efficiency in mitigating membrane fouling. The hydrodynamic parameters (including particle velocity, particle concentration, particle momentum and extent of segregation) of the fluidized particles numerically obtained via a Two-Fluid Model (TFM) were correlated with the fouling rate experimentally obtained. Three bidisperse mixtures with the same mean particle diameter (namely, 1 mm) but different particle diameter ratios (namely, $d_{p2}/d_{p1} = 1.22 - 1.86$) were studied over a range of superficial water velocities. The key highlights are that (i) the fouling rate decreased (i.e., fouling mitigation was more effective) as the particle diameter ratio increased, which indicates the benefits of fluidizing multi-sized particle mixtures over monodisperse ones, (ii) the fouling rate

($dTMP/dt$) was positively correlated to the mixing index (M), which implies that segregated systems were more effective at mitigating membrane fouling, and (iii) the effect of bidispersity was more significant at the lower power input, which suggests that the advantages of fluidizing such bidisperse mixtures should only be harnessed at moderate power but not higher ones.

Keywords: anaerobic fluidized bed membrane bioreactor; bidisperse; two-fluid model; membrane fouling mitigation; hydrodynamics

6.1. Introduction

The fluidization of millimeter-sized granular activated carbon (GAC) particles as an energy-efficient means of mitigating membrane fouling was first proven practically feasible in a continuous anaerobic fluidized-bed membrane bio-reactor (AnFMBR) operated over 120 days [1]. Moreover, the energy consumption was reported to be significantly less than other submerged bioreactors using conventional gas sparging for fouling control. Considerable traction has been gained on understanding and improving the use of fluidized particles to mitigate fouling [2, 3]. Previous studies on particle fluidization inside AnFMBR focused on the impact of particle properties (e.g., type [4, 5], size [6-9]), foulant [1, 10-12] or process parameters (e.g., inlet superficial water velocity [8, 9, 13], pretreatment process [14-18], membrane type [19]) on fouling mitigation process. Although the particles fluidized are inevitably polydisperse (i.e., a range of particle sizes), the impact of the extent of polydispersity on the effectiveness of membrane fouling mitigation is unknown. This underlies the goal of the current study, which focuses on bidisperse or binary size systems (i.e., fluidized particles of two distinctly different sizes). Notably, size-segregation and inter-species

momentum transfer are characteristics unique to non-monodisperse systems, and thereby warrant more understanding of the attendant impact and potential benefits.

Extensive experimental studies on bidisperse solid-liquid fluidized bed systems have been carried out previously. The average expanded bed volume fraction was found to decrease as the mass fraction of the smaller constituent in the bidisperse mixture increased [20]. Epstein et al. [21] subsequently found that, when the terminal velocity ratio was 1.1 – 2.1 and particle Reynold number was 0.02 - 2250, binary mixtures with constituents differing in size, density, or both size and density exhibited expanded bed heights equalling that of the sum of the expanded bed heights of each constituent fluidized separately. The reduced bulk density of the binary mixtures was found to be a useful parameter to predict the degree of segregation [22], and larger differences in the bulk density of the constituents tended to increase the segregation tendency with the constituent with the larger bulk density segregating preferentially to the bottom of the reactor [23]. Segregation in a binary mixture has been attributed to the axial particle convection due to the relative difference of the terminal settling velocities of the constituents, while particle mixing has been attributed to particle diffusion due to the local solid concentration gradient [24, 25]. For bidisperse mixtures, past studies [20, 26, 27] have found that a critical particle diameter ratio exists that demarcates partial and complete segregation. Juma and Richardson [28] found that both the diffusion coefficients of the constituents of bidisperse mixture and axial segregation increased as liquid viscosity decreased or superficial liquid velocity increased or particle diameter ratio increased. They attributed such behaviors to particle circulation within the expanded particle bed, which increased particle convection (i.e., particle segregation) more than particle diffusion (i.e., mixing tendency) [28]. This indicates that operating conditions may exacerbate particle segregation or mixing, depending on the relative

magnitudes of particle diffusion and convection within the system. This is in line with the findings of Wakeman and Stopp [29], who reported that particle segregation increased with superficial liquid velocity. However, semi-empirical equations developed to predict particle segregation in solid-liquid fluidization [24-26, 28, 30-41] have been found to have limited application beyond the scope tested [39, 42], particularly due to the lack of mechanistic understanding on the impact of local turbulence [43], inter-particle collisions [44-46], and particle polydispersity [47]. More recent studies [42, 48-52] have proposed stochastic-based segregation-mixing models, which have wider application but require robust datasets of local particle velocity and concentration over a range of superficial liquid velocities. The numerical method is preferred to provide such data, which otherwise is cumbersome and time-consuming to obtain experimentally [43] (e.g., via particle tracking [32], or pressure gradient measurement [28]). Past numerical studies of solid-liquid fluidization of binary mixtures have focussed on evaluating inter-particle interactions (e.g., cohesive forces, collisions) [45, 53], particle dispersion [43], particle bed expansion [54], drag correlation [55], layer inversion [56, 57], and heat transfer [58].

The studies to date have collectively provided important insights into the fluidization of binary mixtures, and our earlier studies have shed some light on the relationship between the hydrodynamics of monodisperse particles and the extent of fouling mitigation in membrane filtration [8, 9, 13, 59], but the question that remains unaddressed is the benefits or lack thereof of bidisperse mixtures (i.e., particles of two different sizes) for membrane fouling mitigation. Therefore, the overarching goal of the current study was to understand if bidisperse granular activated carbon (GAC) mixtures can mitigate membrane fouling more energy-efficiently. Three bidisperse mixtures with the same mean particle diameter but different constituent particle diameter ratios were

numerically investigated via the two-fluid model (TFM) over a range of superficial liquid velocities. The specific goals were (i) describe the hydrodynamics of the fluidized binary mixtures, (ii) correlate the hydrodynamics with the extent of membrane fouling mitigation, and (iii) determine the energy-efficiency of each bidisperse mixture in mitigating membrane fouling.

6.2. Experimental and Simulation Methods

6.2.1. Experiments for investigating membrane fouling

The fluidized bed membrane reactor (Fig. 6.1a) used for the experiments was cylindrical with a diameter and height of 0.1 m and 0.47 m, respectively. The initial bed height (h_0) was kept consistently at 0.1 m, which was twice that in the simulations because of the twofold difference in reactor diameter [60]. The experimental setup and protocol has been described in detail in Wang et al. [13], and given in brief here. The membrane module had a width, height, and depth of respectively 80 mm, 120 mm and 15 mm, and the lower end was 10 cm above the distributor. Two flat-sheet polyvinylidene fluoride (PVDF) membranes (Merck Millipore Ltd) with a nominal pore size of 0.22 μm and total effective area of 0.0081 m^2 were attached to the back and front faces of the membrane module. The feed was a suspension of 2 g/L bentonite (Sigma-Aldrich) of mean size $5.4 \pm 0.11 \mu\text{m}$ [9]. Three bidisperse (i.e., two different particle diameters) mixtures of GAC particles (particle density (ρ_p) of 1500 kg/m^3) with the same arithmetic mean particle diameter (d_{pm}) of 1 mm, and particle diameter ratios (d_{p2}/d_{p1}) of 1.22, 1.50 and 1.86, were investigated. Specifically, the three particle diameter pairs were 0.9 and 1.1 mm, 0.8 and 1.2 mm, and 0.7 and 1.3 mm, and the two constituents were present at the same mass ratio. Due to limitations of the centrifugal pump (Cole-Parmer) used, the maximum superficial liquid velocity (U_l) possible

experimentally was 0.013 m/s. Superficial liquid velocity was measured using a Cole-Parmer variable area flow-meter (1.5 – 20 LPM). The experiments were all operated at a constant permeate flux of 30 L/m²/h by a suction pump (peristaltic pump Masterflex Cole-Parmer 7528-10, 6 – 600rpm), and the trans-membrane pressure (TMP) was measured by a pressure transmitter (Cole-Parmer, 0 – 1 psi) and recorded every 20 s via LabView (National Instruments).

6.2.2. TFM simulations for investigating the hydrodynamics of the fluidized bidisperse GAC

The numerical study evaluated the fluidization of bidisperse (i.e., two different particle diameters) GAC particles in water using the Two-Fluid Model (TFM) on the open-source platform of Multiphase Flow with interphase eXchange (MFiX) 2016-1 developed by National Energy Technology Laboratory (NETL) [61]. Fig. 6.1b shows the simulation domain of the three-dimensional (3D) cylindrical anaerobic fluidized membrane bioreactor (AnFMBR) with diameter and height of 0.05 m and 0.30 m, respectively. The initial bed height (h_0) was consistently set at 0.05 m. It should be noted that this setup was geometrically similar but smaller than that used for the experiments (Fig. 6.1a), which was similar to an earlier experimental study [13], to reduce the computational duration. The diameter and initial bed height of the simulated reactor were both half that of the experimental one [60].

Fig. 6.1. (a) Experimental fluidized bed - membrane reactor; (b) Simulation domain of the 3D AnFMBR.

The TFM equations used have been presented in our previous study on monodisperse GAC particles [8], and are shown in the Appendix (Table 6.A1). Tables 6.1 and 6.2 list respectively the simulation parameters and operating conditions employed in this study. The fluidized particles of interest were granular activated carbon (GAC) with particle density (ρ_p) of 1500 kg/m^3 and particle diameters ranging between $0.7 - 1.3 \text{ mm}$, while the liquid phase was water with a density (ρ_w) of 1000 kg/m^3 (Table 6.1). The same bidisperse GAC mixtures (Table 6.2) were used in both the experiments and the simulations. For each bidisperse mixture, the two species were present at the same volume ratio, and the initial condition was a well-mixed system. To ensure negligible wall effect [62], the ratio of the column diameter and largest GAC particle diameter was larger than 10. The characteristic minimum fluidization velocities (U_{mf}) of each particle are listed in Table 6.2, while the terminal velocities (U_t) are given in the

appendix (Table 6.A2) to indicate the GAC particles were not entrained. The minimum fluidization velocities of the binary mixtures ($U_{mf,b}$) listed in Table 6.2 was calculated by [63]:

$$U_{mf,b} = U_{mf,1} \left(\frac{U_{mf,2}}{U_{mf,1}} \right)^{\overline{m}_{p2}^2} \quad (1)$$

where $U_{mf,1}$ and $U_{mf,2}$ represent the minimum fluidization velocity of respectively the smaller and larger constituent, and $\overline{m}_{p2}$ represents the average mass fraction of the larger constituent at the initial condition (i.e., 0.5). Also listed in Table 6.2 are the superficial liquid velocities (U_l) assessed in the range of $U_l/U_{mf,b}$ values of 2 – 11, the lower bound of which enabled some expansion of the fluidized bed and the upper bound of which prevented entrainment of the smaller constituent (terminal velocities listed in Table 6.A2).

Table 6.1. Simulation parameters used for bidisperse GAC study in 3D AnFMBR.

Parameter	Unit	Magnitude
Simulation time	s	80 (only the final 60 s is used for steady-state data analysis)
Time step	s	0.0001
Total number of grids	-	7750
Water Properties		
Water density (ρ_w)	kg/m ³	1000
Water viscosity (μ_w)	kg/m/s	0.001
Turbulent length scale (l)	m	0.00336
Maximum turbulent viscosity (μ_{wt})	kg/m/s	100

GAC properties		
GAC particle density (ρ_p)	kg/m ³	1500
GAC particle diameters (d_p) in the bidisperse mixtures	mm	0.9 and 1.1 ($d_{p2}/d_{p1} = 1.22$) 0.8 and 1.2 ($d_{p2}/d_{p1} = 1.50$) 0.7 and 1.3 ($d_{p2}/d_{p1} = 1.86$)
Maximum packing of bidisperse particles	-	Fedors-Landel equation [64]
Initial slope of segregation [65]	-	0.3
Particle-particle coefficient of friction (μ_k)	-	0.3
Particle-particle restitution coefficient (e_p)	-	0.99
Particle-wall restitution coefficient (e_r)	-	0.99
Angle of internal friction ($\emptyset$)	°	30
Angle of internal friction at wall ($\emptyset_r$)	°	11.31
Specularity coefficient at wall ($\emptyset'$) [66]	-	0.0001
Initial Conditions		
Particle Bed		
Bed height (h_0)	m	0.05
Mass fraction of particles corresponding to smaller constituent (m_{p1})	-	0.5
Mass fraction of particles corresponding to larger constituent (m_{p2})	-	0.5
Volume-based void fraction ($\varepsilon_{l,b}$)	-	0.5
Volume fraction of smaller constituent (ε_{p1})	-	0.25
Volume fraction of larger constituent (ε_{p2})	-	0.25

Freeboard		
Void fraction ($\varepsilon_{l,f}$)	-	1 (i.e., no GAC particles)
Boundary Conditions		
Bottom ($h/H = 0$): uniform superficial liquid velocity (U_l)	m/s	$5.14 \times 10^{-3} - 3.10 \times 10^{-2}$
Top ($h/H = 1$): uniform water pressure	Pa	101325
Sides ($r/R = -1$ and 1): partial slip for particle-wall interaction	-	Johnson-Jackson boundary conditions [67]

Table 6.2. Operating conditions used for bidisperse GAC study in 3D AnFMBR.

d_p (mm)		U_{mf} [68] (m/s)		d_{p2} $/d_{p1}$	$U_{mf,b}$ [63] (m/s)	U_l (m/s)				
d_{p1}	d_{p2}	$U_{mf,1}$	$U_{mf,2}$			$\frac{U_l}{U_{mf,b}} =$ 2	$\frac{U_l}{U_{mf,b}} =$ 4	$\frac{U_l}{U_{mf,b}} =$ 6	$\frac{U_l}{U_{mf,b}} =$ 9	$\frac{U_l}{U_{mf,b}} =$ 11
0.9	1.1	0.0023	0.0034	1.22	0.0028	0.0056	0.0113	0.0169	0.0253	0.0310
0.8	1.2	0.0019	0.0040	1.50	0.0027	0.0054	0.0109	0.0163	0.0245	0.0299
0.7	1.3	0.0014	0.0046	1.86	0.0026	0.0051	0.0103	0.0154	0.0231	0.0282

6.3. Results and Discussion

6.3.1. Fouling trends from experiment

Armed with the goal of understanding whether bidisperse mixtures are more energy-efficient than the monodisperse counterpart, filtration experiments were carried out using fluidized bidisperse GAC particles to correlate the hydrodynamics parameters numerically obtained with the extent of membrane fouling mitigation. Fig. 6.2 plots

trans-membrane pressure (TMP) versus time for the fluidized bidisperse GAC with particle diameter ratios (d_{p2}/d_{p1}) of 1.22 and 1.86, and superficial liquid velocity ratios ($U_l/U_{mf,b}$) in the range of 2 - 4. Two observations are worth noting. Firstly, expectedly, the rates of TMP increase were lower for the higher superficial liquid velocity ratios ($U_l/U_{mf,b}$) regardless of particle diameter ratios (d_{p2}/d_{p1}). This is because the increased movement of the fluidized GAC (e.g., particle velocity and momentum) increased the mechanical scouring of the membrane. Secondly, although relatively similar pump energy was expended to fluidize both bidisperse mixtures at each superficial liquid velocity ratio ($U_l/U_{mf,b}$), the one with the larger particle diameter ratio (d_{p2}/d_{p1}) of 1.86 gave slower rates of TMP increase and thereby was more effective in mitigating membrane fouling.

Fig. 6.2. Trans-membrane pressure (TMP) versus time (t) for fluidized bidisperse GAC mixtures with particle diameter ratios (d_{p2}/d_{p1}) of (a) 1.22 and (b) 1.86.

6.3.2. Particle hydrodynamics from TFM simulation

- **Model validation**

Fig. 6.3 presents the TFM-computed radial-averaged bed void fraction ($\varepsilon_{l,b}$) at the axial position corresponding to the initial bed height (h_0) of 0.05 m in the time (t) interval between 0 and 80 s for all three bidisperse mixtures at superficial liquid velocity

ratio ($U_l/U_{mf,b}$) values of 2 and 11. Similar trends of radial-averaged bed voidage fractions ($\varepsilon_{l,b}$) versus time were found for other heights within the expanded fluidized bed. Three observations warrant mentioning. Firstly, the final radial-averaged bed voidage fractions ($\varepsilon_{l,b}$) increased as the particle diameter ratio (d_{p2}/d_{p1}) decreased or as the superficial liquid velocity ratio ($U_l/U_{mf,b}$) increased. Secondly, an increase in the superficial liquid velocity ratio ($U_l/U_{mf,b}$) increased the time needed to achieve steady-state (i.e., the plateauing of $\varepsilon_{l,b}$ with time), while an increase in the particle diameter ratio (d_{p2}/d_{p1}) decreased it. Finally, because the bed void fractions ($\varepsilon_{l,b}$) became constant with time for all cases after 20 s, the evaluation of system parameters in the following sections will be based on the temporal average in the time interval of $t = 21 - 80$ s.

Fig. 6.3. Evolution of the radially averaged bed void fraction ($\varepsilon_{l,b}$) of the fluidized bidisperse GAC particles at a bed height (h) of 5 cm.

The spatially averaged bed void fractions ($\varepsilon_{l,b}$) at steady state condition numerically obtained were compared against the experimental data for the bidisperse mixtures with particle diameter ratios (d_{p2}/d_{p1}) of 1.22 and 1.86 and at superficial liquid velocities (U_l) between 0.005 and 0.011 m/s, as displayed in Fig. 6.4. Specifically, the $\varepsilon_{l,b}$ values were experimentally obtained by (1) quantifying the particle volume fraction of the fixed bed by measuring the volume of water needed to fill the reactor to the height of the fixed particle bed, then (2) quantifying the expanded bed height, which gives the

expanded bed volume. The upper bound of superficial liquid velocity (U_l) assessed was limited by the experimental data available due to limitation of the centrifugal pump used. Fig. 6.4 shows that numerical data of the spatially-averaged expanded bed void fraction ($\varepsilon_{l,b}$) agree very well with experimental data for both the lowest and highest particle diameter ratios (d_{p2}/d_{p1}) of the bidisperse mixtures investigated.

Fig. 6.4. Comparison between simulation and experimental results of spatially averaged bed void fraction ($\varepsilon_{l,b}$) for bidisperse GAC particles with particle ratios (d_{p2}/d_{p1}) of 1.22 and 1.86, and in the superficial liquid velocity (U_l) range of 0.005 and 0.011 m/s.

- **Hydrodynamics of fluidized bidisperse GAC**

The effectiveness of mechanical scouring by the fluidized GAC to mitigate membrane fouling depends on expanded particle bed height which is tied to the membrane area scoured, and particle concentration and velocity which affect the extent of scouring. Accordingly, the numerical study here evaluates expanded bed height, the particle velocity distribution profiles, the distribution of the constituent particles in the bidisperse mixture, and particle momentum. Specifically, armed with the understanding that the hydrodynamics of fluidized monodisperse GAC particles correlate well with the

extent of membrane fouling mitigation [8, 9], this section aimed at evaluating the impact of the particle diameter ratio of bidisperse mixtures on the fluidization characteristics.

Fig. 6.5 displays the TFM results for the expanded bed height and axial bulk density (ρ_b) distribution of the smaller and larger constituents of the bidisperse GAC mixtures with particle diameter ratios (d_{p2}/d_{p1}) of 1.22 and 1.86 at the superficial liquid velocity ratios ($U_l/U_{mf,b}$) ranging between 2 and 11.

Fig. 6.5. Expanded bed height (h) and contour plots of the bulk density (ρ_b) of the smaller and larger constituents at $U_l/U_{mf,b} = 2 - 11$ for the fluidized bidisperse GAC mixtures with particle diameter ratios (d_{p2}/d_{p1}) of (a) 1.22 and (b) 1.86.

Note that the expanded bed height is reflected in the height attained by the smaller constituents (left column of Fig. 6.5) due to size-segregation. Two findings are

discussed here. Firstly, the size-segregation was such that (i) the smaller constituents preferentially segregated to higher up in the fluidized bed, and (ii) the extent of segregation increased with superficial liquid velocity ratio ($U_l/U_{mf,b}$) and with particle diameter ratio (d_{p2}/d_{p1}), which agrees with earlier experimental studies [21, 28]. At the lowest superficial liquid velocity ratio ($U_l/U_{mf,b}$) of 2, both constituents were found throughout the most of the bed for the lower particle diameter ratio (d_{p2}/d_{p1}) of 1.22 (Fig. 6.5a), but the segregation was expectedly more extensive for the higher particle diameter ratio (d_{p2}/d_{p1}) such that only the larger constituents were found lower in the bed (Fig. 6.4b). On the other hand, at the higher superficial liquid velocity ratios ($U_l/U_{mf,b}$) of 9 and 11, the segregation was perfect for both particle diameter ratios (d_{p2}/d_{p1}) of 1.22 and 1.86, specifically in that only the larger constituents were found lower in the bed while only the smaller ones higher in the bed. Secondly, as the superficial liquid velocity ratio ($U_l/U_{mf,b}$) increased, the fluidized bed height increased while the width decreased, whereas the change in particle diameter ratio (d_{p2}/d_{p1}) did not affect the fluidized bed height and width. The decrease in width agrees with a previous study [69].

Figs. 6.6 – 6.9 provide insights from the TFM into the hydrodynamics of each particle constituent in the three bidisperse mixtures (namely, with particle diameter ratios (d_{p2}/d_{p1}) of 1.22, 1.50, and 1.86) throughout the reactor at three superficial liquid velocity ratios ($U_l/U_{mf,b}$). Figs. 6.6, 6.7, 6.8 and 6.9 contains 9 sub-figures each, and reflect respectively the average particle velocity ($U_{p,ave}$), particle mass fraction (m_p), volumetric fraction (ε_p) and particle momentum (p_p). In each figure, each column represents a superficial liquid velocity ratio ($U_l/U_{mf,b}$; namely, 2, 6 and 11), while each of the three row represents a dimensionless height (h/H , where h is the height assessed and H is total expanded fluidized bed height). Each sub-figure plots the hydrodynamic parameter versus the dimensionless reactor radius (r/R , where r is the radial position

assessed and R is the reactor radius of 0.05 m, and $r/R = 0$ represents the reactor center), and contains six trends for the two constituents in each of the three bidisperse mixtures.

Fig. 6.6 portrays the average particle velocity ($U_{p,ave}$) trends, since particle velocities are closely tied to the extent of fouling mitigation [8, 9]. It should be noted that the average particle velocity ($U_{p,ave}$) magnitudes were dominated by the vertical velocity component, which agrees with our previous study on monodisperse particles [8]. Four findings are highlighted. Firstly, the average particle velocities ($U_{p,ave}$) were more uniform radially at the lower superficial liquid velocity ratio ($U_l / U_{mf,b}$) of 2 (Fig. 6.6a), but significantly higher near the walls at the higher superficial liquid velocity ratios ($U_l / U_{mf,b}$) of 6 - 11 (Figs. 6.6b and c). This implies that the fouling mitigation across the membrane was more uniform at lower superficial liquid velocity ratios ($U_l / U_{mf,b}$). Secondly, the size-segregation effects were greater at the higher superficial liquid velocity ratios ($U_l / U_{mf,b}$) of 6 - 11 (Figs. 6.6b and c), as evident in the lack of larger particles at $h/H = 0.8$, which agrees with Fig. 6.5. This indicates that only the smaller particles were available to mitigate membrane fouling higher in the reactor at the higher superficial liquid velocity ratios ($U_l / U_{mf,b}$). Thirdly, among the bidisperse mixtures, no consistent correlation existed between average particle velocity ($U_{p,ave}$) and particle diameter ratio (d_{p2}/d_{p1}). This implies that the particle diameter ratio (d_{p2}/d_{p1}) had a weak influence on the average particle velocity ($U_{p,ave}$). Fourthly, between the two constituents in the bidisperse mixtures, the smaller constituent generally had greater average particle velocities ($U_{p,ave}$) than the larger. This is due to the greater mobility conferred by the greater magnitude of $(U_l - U_{mf,b})$, which underlies why smaller particles have been reported in our earlier work to be more energy-efficient at mitigating fouling [8].

Fig. 6.6. Local average particle velocity ($U_{p,ave}$) with respect to reactor radius (r/R) for three bidisperse GAC mixtures ($d_{p2}/d_{p1} = 1.22, 1.50$ and 1.86) at three heights ($h/H = 0.2, 0.5$ and 0.8) for three superficial liquid velocity ratios ($U_l/U_{mf,b}$) of (a) 2, (b) 6, and (c) 11.

Fig. 6.7 displays the particle mass fraction (m_p) trends, which reflect the size-segregation effects, which in turn influences the local mix of constituents that scour the corresponding portion of the membrane. The number-based trends are depicted in Fig. 6.A1. The size-segregation trends generally agree with Juma and Richardson [28]. Three

observations are noted. Firstly, the size-segregation effects increased as the superficial liquid velocity ratio ($U_l/U_{mf,b}$) increased. At the higher superficial liquid velocity ratios ($U_l/U_{mf,b}$; Figs. 6.7b and c), it is clear that (i) only the smaller constituent was present at the highest height ($h/H = 0.8$), and (ii) the smaller constituent was dominant near the wall ($r/R \sim 1$) at the lowest height ($h/H = 0.2$). Secondly, the extent of segregation was closely tied to height (h/H). At the lower superficial liquid velocity ratio ($U_l/U_{mf,b}$; Fig. 6.7a), the radial particle mass fraction (m_p) trends were quite uniform at the lowest height ($h/H = 0.2$), but the smaller constituent preferentially segregated to the wall at the higher heights ($h/H \geq 0.5$). On the other hand, at the higher superficial liquid velocity ratios ($U_l/U_{mf,b}$; Figs. 6.7b - c), the radial segregation was clear at the lower heights ($h/H = 0.2$), while only the smaller constituents were present at the highest height ($h/H = 0.8$). Thirdly, the extent of segregation increased with the particle diameter ratio (d_{p2}/d_{p1}) of the bidisperse mixture. Although the overall mass fraction (m_p) of each constituent was constant at 0.5, the differences in the local mass fraction values of the smaller (m_{p1}) and larger (m_{p2}) constituents increased as the particle diameter ratio (d_{p2}/d_{p1}) increased.

Fig. 6.7. Local particle mass fraction (m_p) with respect to reactor radius (r/R) for three bidisperse GAC mixtures ($d_{p2}/d_{p1} = 1.22, 1.50$ and 1.86) at three heights ($h/H = 0.2, 0.5$ and 0.8) for three superficial liquid velocity ratios ($U_l/U_{mf,b}$) of (a) 2, (b) 6, and (c)

11.

Fig. 6.8 displays the volumetric fraction of each constituent (ε_p) in the bidisperse mixtures. Whereas Fig. 6.7 considers only the particles without the water phase, Fig. 6.8 accounts for both the water phase and the particles, specifically in that the sum of the

volumetric fractions of both constituents ($\varepsilon_{p1} + \varepsilon_{p2}$) indicates the local concentration of the particles. The overall particle concentration ($\varepsilon_{p1} + \varepsilon_{p2}$), which has been reported to be tied to the extent of membrane fouling mitigation [8, 13, 59], has similar trends to that of the monodisperse system previously studied [8]. Four observations are described as follows. Firstly, the local particle concentration ($\varepsilon_{p1} + \varepsilon_{p2}$) decreased as height or superficial liquid velocity ratio ($U_l/U_{mf,b}$) increased. Secondly, the particle concentration ($\varepsilon_{p1} + \varepsilon_{p2}$) was more uniform with respect to both height and radial position at the lowest superficial liquid velocity ratio ($U_l/U_{mf,b}$; Fig. 6.8a), and the non-uniformity increased as the superficial liquid velocity ratio ($U_l/U_{mf,b}$) increased. Thirdly, the larger constituents were clearly dominant at the lower ($h/H = 0.2$) height, and the corresponding volumetric fractions (ε_{p2}) were higher than that of the smaller constituents dominating at the higher ($h/H = 0.8$) heights. Fourthly, the effect of varying bidispersity on local particle concentration ($\varepsilon_{p1} + \varepsilon_{p2}$) depended on the superficial liquid velocity ratio ($U_l/U_{mf,b}$). At the lower superficial liquid velocity ratio ($U_l/U_{mf,b}$; Fig. 6.8a), varying bidispersity had negligible impact on the local particle concentration ($\varepsilon_{p1} + \varepsilon_{p2}$). On the other hand, at the higher superficial liquid velocity ratios ($U_l/U_{mf,b}$; Figs. 6.8b - c), the bidisperse mixture with the greatest particle diameter ratio (d_{p2}/d_{p1}) gave the highest local particle concentration ($\varepsilon_{p1} + \varepsilon_{p2}$) at the lowest height ($h/H = 0.2$), whereas that with the least particle diameter ratio (d_{p2}/d_{p1}) gave the highest local particle concentration ($\varepsilon_{p1} + \varepsilon_{p2}$) at the highest height ($h/H = 0.8$).

Fig. 6.8. Local particle volume fraction (ϵ_p) with respect to reactor radius (r/R) for three bidisperse GAC mixtures ($d_{p2}/d_{p1} = 1.22, 1.50$ and 1.86) at three heights ($h/H = 0.2, 0.5$, and 0.8) for three superficial liquid velocity ratios ($U_l/U_{mf,b}$) of (a) 2, (b) 6, and (c) 11.

Fig. 6.9 presents the particle momentum (p_p) trends, which have been reported to be correlated most positively with the extent of membrane fouling mitigation [8, 9].

Specifically, the local particle momentum (p_p) is the product of the local mass of particles (M_p) and local average particle velocity ($U_{p,ave}$), as described in equation (2):

$$p_p = (M_{p1} \cdot U_{p1,ave}) + (M_{p2} \cdot U_{p2,ave}) \quad (2)$$

where the subscripts 1 and 2 represent the smaller and larger constituents, respectively. The particle momentum trends of each constituent are shown in Fig. 6.A2. Four highlights in Fig. 6.9 are noted. Firstly, the particle momentum (p_p) was highly non-uniform across the membrane both with respect to height and radial position, which agrees with that for the monodisperse system [8]. Secondly, the highest particle momentum magnitudes were exhibited by the bidisperse mixture with the greatest particle diameter ratio (d_{p2}/d_{p1}) at the lowest height (h/H) and lowest superficial liquid velocity ratio ($U_l/U_{mf,b}$), which is due to both higher mass fraction (m_p ; Fig. 6.7) and average velocity ($U_{p,ave}$; Fig. 6.6) of the larger constituent. Thirdly, the particle momentum (p_p) was more uniform with height for the highest superficial liquid velocity ratio ($U_l/U_{mf,b}$; Fig. 6.9c). Fourthly, a direct correlation between bidispersity and particle momentum is not clear in Fig. 6.9. To further understand the relationship, Fig. 6.10 plots the spatially averaged particle momentum (p) with respect to the superficial liquid velocity ratio ($U_l/U_{mf,b}$). As expected, particle momentum (p) generally increased with the superficial liquid velocity ratio ($U_l/U_{mf,b}$). However, whereas the particle momentum (p) of the bidisperse mixture with the least particle diameter ratio (d_{p2}/d_{p1}) increased monotonically with superficial liquid velocity ratio ($U_l/U_{mf,b}$), that of the bidisperse mixtures with greater particle diameter ratios (d_{p2}/d_{p1}) trended non-monotonically with superficial liquid velocity ratio ($U_l/U_{mf,b}$). The latter is likely due to momentum transfer between the different constituent types at various extents of size-segregation.

Fig. 6.9. Local particle momentum (p_p) with respect to reactor radius (r/R) for three bidisperse GAC mixtures ($d_{p2}/d_{p1} = 1.22, 1.50$, and 1.86) at three heights ($h/H = 0.2, 0.5$, and 0.8) for three superficial liquid velocity ratios ($U_l/U_{mf,b}$) of (a) 2, (b) 6, and (c)

Fig. 6.10. Particle momentum (p) versus superficial water velocity ratios ($U_l/U_{mf,b}$) for bidisperse mixtures with different particle diameter ratios (d_{p2}/d_{p1}).

Table 6.3 summarizes the key observations from Figs. 6.5 – 6.10.

Table 6.3. Impact of increases in the superficial liquid velocity ratio ($U_l/U_{mf,b}$) and particle diameter ratio (d_{p2}/d_{p1}) on the hydrodynamics of fluidized bidisperse GAC in the AnFMBR.

	Increase in Superficial Liquid Velocity Ratio ($U_l/U_{mf,b}$)	Increase in Particle Diameter Ratio (d_{p2}/d_{p1})
Membrane area scoured	<ul style="list-style-type: none"> Expanded bed height increased Expanded bed width decreased Scouring area increased 	No effect.
Average particle velocity ($U_{p,ave}$)	<ul style="list-style-type: none"> Particle recirculation increased $U_{p,ave}$ near the wall increased more than that at the centre 	<ul style="list-style-type: none"> Low $U_l/U_{mf,b}$: no clear correlation. High $U_l/U_{mf,b}$: no clear correlation at annulus,

[axial particle velocity dominant]	<ul style="list-style-type: none"> • $U_{p,ave}$ of smaller constituent increased more than that of larger constituent. 	$U_{p,ave}$ decreased with d_{p2}/d_{p1} near the wall
Particle segregation [axial segregation more dominant than radial segregation]	<ul style="list-style-type: none"> • Axial segregation increased: larger particles tended to occupy the bottom • Radial segregation increased: larger particles tended to occupy the centre 	Axial and radial particle segregation increased
Particle concentration ($\varepsilon_{p1} + \varepsilon_{p2}$)	<ul style="list-style-type: none"> • Particle concentration decreased • Axial and radial uniformities of particle concentration decreased 	<ul style="list-style-type: none"> • Low $U_l/U_{mf,b}$: no effect • High $U_l/U_{mf,b}$: maximum local particle concentration shifted from top to bottom of column.
Particle momentum (p_p)	Particle momentum increased	No clear correlation

6.3.3. Relationship between fluidization hydrodynamics and fouling mitigation efficiency

Fig. 6.11 correlates the experimental fouling rate ($dTMP/dt$) with respect to the TFM calculated mass-based average particle velocity (U_p ; Fig. 6.11a), mass-based average particle concentration ($1 - \varepsilon_{l,b}$; Fig. 6.11b), mass-based average particle

momentum (p ; Fig. 6.11c), power required (Fig. 6.11d), and mixing index (M ; Fig. 6.11e), and for the two binary mixtures with particle diameter ratios (d_{p2}/d_{p1}) of 1.22 and 1.86. The power required was calculated by:

$$\text{Power} = U_l \cdot A \cdot \Delta P \quad (3)$$

where U_l is the inlet superficial liquid velocity, A is the cross-sectional area of the reactor, and ΔP is the pressure drop across the expanded particle bed. The mixing index (M) was calculated by:

$$M = \frac{m_{p2,h/H=0.9}}{0.5} \quad (4)$$

where $m_{p2,h/H=0.9}$ is the mass fraction of the larger constituent at $h/H = 0.9$, and 0.5 is the mass fraction of the larger constituent in the entire bed. Mixing index (M) values of 0 and 1 represent complete segregation and complete mixing, respectively. The error bars represent the span of the dTMP/dt values obtained from three repeated tests. Figs. 6.11a – c indicates that the fouling rate (dTMP/dt) decreased (i.e., fouling mitigation was more effective) as either the average particle velocity (U_p) increased (Fig. 6.11a) or particle concentration decreased (Fig. 6.11b) or particle momentum (p) increased (Fig. 6.11c) or power increased (Fig. 6.11d), which agrees with previous findings [8, 9]. Furthermore, Fig. 6.11e shows that the fouling rate (dTMP/dt) was positively correlated to the mixing index (M), which implies that segregated systems were more effective at mitigating membrane fouling. Regarding the impact of bidispersity on the extent of fouling mitigation, the effect of particle diameter ratio (d_{p2}/d_{p1}) was only significant for the relationships with particle concentration ($1 - \varepsilon_{l,b}$; Fig. 6.11b) and power (Fig. 6.11d). Notably, at the lower power input, the dTMP/dt value was distinctly lower for the bidisperse mixture with the greater particle diameter ratio (d_{p2}/d_{p1}), which implies that the fouling mitigation was more effective. As the power input increased, the fouling

mitigation effectiveness became more similar between the two bidisperse mixtures. This signifies the benefits of using increased differential between the two particle diameters (d_{p2}/d_{p1}) in a bidisperse mixture to more efficiently mitigate fouling at a moderate power. This also suggests the advantage of fluidizing multi-sized GAC particles over monodisperse ones for more effective membrane fouling mitigation, at least in part owing to inter-particle collisions between smaller and larger constituents in increasing the scouring efficiency of the larger constituents.

Fig. 6.11. Fouling rate ($dTMP/dt$) versus (a) mass-based average particle velocity ($U_{p,ave}$), (b) mass-based average particle concentration ($1 - \epsilon_{l,b}$), (c) mass-based average particle momentum (p), (d) power required, (e) mixing index (M).

6.4. Conclusions

Armed with the question of whether the fluidization of bidisperse GAC mixtures can be more energy-efficient relative to monodisperse ones in mitigating membrane fouling, the current study evaluated the hydrodynamics of the fluidized particles numerically via a Two-Fluid Model (TFM) and the fouling rate experimentally. Three bidisperse mixtures with the same mean particle diameter but different particle diameter ratios (d_{p2}/d_{p1}) between 1.22 and 1.86 were studied under a range of superficial water velocity ratios ($U_l/U_{mf,b}$) between 2 and 11. It was assumed that the lowest particle diameter ratio (d_{p2}/d_{p1}) of 1.22 mimicked the monodisperse system most, hence the monodisperse system (whose relationship between hydrodynamics and fouling mitigation efficacy have been reported [8, 9, 13, 59]) was not studied here. Hydrodynamics characteristics like particle velocity ($U_{p,ave}$), particle concentration ($1 - \varepsilon_{l,b}$), particle momentum (p) and extent of segregation (M) were correlated with the rate of change of trans-membrane pressure ($dTMP/dt$).

Regarding the impact of bidisperse mixtures, it was found experimentally that the fouling rate ($dTMP/dt$) decreased (i.e., fouling mitigation was more effective) as the particle diameter ratio (d_{p2}/d_{p1}) increased. The numerical results indicate the general lack of direct correlations between particle diameter ratio (d_{p2}/d_{p1}) and the trends of either expanded bed height (h), particle velocity ($U_{p,ave}$), local particle concentration ($\varepsilon_{p1} + \varepsilon_{p2}$) or particle momentum (p). Key observations unique to bidisperse mixtures with no monodisperse counterparts are that: (i) the smaller constituent generally had greater average particle velocities ($U_{p,ave}$) than the larger, and preferred the wall and upper regions of the reactor; and (ii) the size-segregation effects increased as the superficial liquid velocity ratio ($U_l/U_{mf,b}$) increased or as the particle diameter ratio (d_{p2}/d_{p1}) increased.

The relationship between fouling rate ($dTMP/dt$) and the fluidization hydrodynamics of the bidisperse mixtures shows expectedly that the fouling rate ($dTMP/dt$) decreased as either the average particle velocity (U_p) increased or particle concentration decreased or particle momentum (p) increased or power increased, which agrees with previous findings [8, 9]. Additional insights are that (i) the fouling rate ($dTMP/dt$) was positively correlated to the mixing index (M), which implies that segregated systems were more effective at mitigating membrane fouling; and (ii) bidispersity was only influential in affecting the relationships between fouling rate ($dTMP/dt$) and particle concentration ($\varepsilon_{p1} + \varepsilon_{p2}$), and between fouling rate ($dTMP/dt$) and power required. More specifically on (ii), at the same particle concentration ($\varepsilon_{p1} + \varepsilon_{p2}$) or power, the bidisperse mixture with the greater particle diameter ratio (d_{p2}/d_{p1}) was more effective at mitigating membrane fouling. Notably, at the lower power input, the $dTMP/dt$ value was distinctly lower for the bidisperse mixture with the greater particle diameter ratio (d_{p2}/d_{p1}), which implies that the fouling mitigation was more effective. As the power input increased, the fouling mitigation effectiveness became more similar between the two bidisperse mixtures. This signifies the benefits of using increased differential between the two particle diameters (d_{p2}/d_{p1}) in a bidisperse mixture to more efficiently mitigate fouling at a moderate power. This also suggests the advantage of fluidizing multi-sized GAC particles over monodisperse ones for more energy-efficient membrane fouling mitigation.

Nomenclature

A	= reactor cross-section area, m^2
Ar	= Archimedes number, –
d^*	= dimensionless particle diameter, –
d_{pm}	= mean particle diameter of bidisperse mixture, mm
d_{p1}	= particle diameter of smaller constituent in bidisperse mixture, mm

d_{p2}	= particle diameter of larger constituent in bidisperse mixture, mm
C_d	= particle drag coefficient, –
D_c	= reactor diameter, m
e_p	= particle-particle restitution coefficient, –
e_r	= particle-wall restitution coefficient, –
g	= gravitational acceleration, m/s ²
g_0	= equilibrium radial distribution function at contact, –
h	= axial position in reactor, m
h_0	= initial particle bed height, m
H	= total expanded bed height, m
H_c	= reactor height, m
J_{wp}	= granular temperature transfer between water and solid, kg/m/s ³
l	= turbulent length scale, m
m_{p1}	= mass fraction of smaller particle inside AnFMBR, –
m_{p2}	= mass fraction of larger particle inside AnFMBR, –
M_p	= local mass of particles, kg
M	= mixing index, –
$\vec{n}$	= unit normal vector from a boundary to particle, –
n_p	= local amount of particle, –
p_p	= local particle momentum, kg.m/s
p_{wp}	= water-solid momentum transfer [61], kg.m/s
$q_{\theta p}$	= diffusive flux of granular temperature, kg/s ³
r	= radial position in reactor, m
R	= reactor radius, m
Re_{mf}	= Reynolds number at minimum fluidization, –
Re_p	= Reynolds number of particle, –
Re_w	= Reynolds number of water, –
$\overline{\overline{S_p}}$	= frictional-kinetic solid stress [70], kg/m/s ²
$\overline{\overline{S_w}}$	= water stress [61], kg/m/s ²
U_*	= dimensionless particle velocity, –
U_l	= inlet superficial water velocity, m/s
$U_{mf,b}$	= minimum fluidization velocity of bidisperse mixture, m/s
$U_{mf,1}$	= minimum fluidization velocity of smaller particle, m/s
$U_{mf,2}$	= minimum fluidization velocity of larger particle, m/s
$U_{p,ave}$	= local average of particle velocity, m/s
U_{t1}	= terminal velocity of smaller particle, m/s
U_{t2}	= terminal velocity of larger particle, m/s

Symbol and Greek Letters

ΔP	= pressure drop across fluidized bed, Pa
ΔV	= differential volume assessed, m ³
β	= interphase drag coefficient, –
$\varepsilon_{l,b}$	= bed void fraction, –
$\varepsilon_{l,f}$	= freeboard void fraction, –
ε_{p1}	= volume fraction of smaller particle, –
ε_{p2}	= volume fraction of larger particle, –
μ_w	= water viscosity, kg/m/s
μ_{wt}	= maximum turbulent viscosity, kg/m/s

θ_p	= granular temperature, m^2/s^2
$\emptyset$	= angle of internal friction, $^\circ$
$\emptyset_r$	= angle of internal friction at wall, $^\circ$
$\emptyset'$	= specularity coefficient at wall, –
ρ_w	= water density, kg/m^3
ρ_p	= particle density, kg/m^3
γ_{θ_p}	= rate of dissipation of granular energy, $\text{kg}/\text{m}/\text{s}^3$

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Appendix

Table 6.A1. CFD equations used for bidisperse GAC study in 3D AnFMBR.

Continuity Equations	
Liquid phase	$\frac{\partial}{\partial t}(\varepsilon_{l,b}\rho_w) + \nabla \cdot (\varepsilon_{l,b}\rho_w \vec{U}_w) = 0$
Solid phase	$\frac{\partial}{\partial t}((1 - \varepsilon_{l,b})\rho_p) + \nabla \cdot ((1 - \varepsilon_{l,b})\rho_p \vec{U}_p) = 0$
Momentum Equations	
Liquid phase	$\frac{\partial}{\partial t}(\varepsilon_{l,b}\rho_w \vec{U}_w) + \nabla \cdot (\varepsilon_{l,b}\rho_w \vec{U}_w \vec{U}_w)$ $= \nabla \cdot \vec{\overline{S}}_w + \varepsilon_{l,b}\rho_w \vec{g} - \varepsilon_{l,b}\nabla P - I_{wp}$
Solid phase	$\frac{\partial}{\partial t}((1 - \varepsilon_{l,b})\rho_p \vec{U}_p) + \nabla \cdot ((1 - \varepsilon_{l,b})\rho_p \vec{U}_p \vec{U}_p)$ $= \nabla \cdot \vec{\overline{S}}_p + (1 - \varepsilon_{l,b})\rho_p \vec{g} - (1 - \varepsilon_{l,b})\nabla P$ $+ I_{wp}$

	where $I_{wp} = \beta(\overrightarrow{U_w} - \overrightarrow{U_p})$
Constitutive Equations	
Interphase drag coefficient [71]	$\beta = 150 \frac{(1-\varepsilon_{l,b})^2 \mu_w}{\varepsilon_{l,b} d_p^2} + 1.75 \frac{(1-\varepsilon_{l,b}) \rho_w \overrightarrow{U_p} - \overrightarrow{U_w} }{d_p}, \text{ for } \varepsilon_{l,b} < 0.8 \text{ [72]}$ $\beta = \frac{3}{4} C_D \frac{(1-\varepsilon_{l,b}) \rho_w \overrightarrow{U_p} - \overrightarrow{U_w} }{d_p} \varepsilon_{l,b}^{-1.65}, \text{ for } \varepsilon_{l,b} \geq 0.8 \text{ [73]}$ <p>where $C_D = \frac{24}{Re_p} (1 + 0.15 Re_p^{0.687}), \text{ for } Re_p \leq 1000$</p> $C_D = 0.44, \text{ for } Re_p > 1000$
Granular temperature [74]	$\frac{3}{2} \left[\frac{\partial}{\partial t} ((1 - \varepsilon_{l,b}) \rho_p \theta_p) + \nabla \cdot ((1 - \varepsilon_{l,b}) \rho_p \theta_p \overrightarrow{U_p}) \right]$ $= [\overline{S_p} : \nabla \overrightarrow{U_p} - \nabla \cdot \overrightarrow{q_{\theta_p}} - \gamma_{\theta_p} + I_{wp}]$ <p>where</p> $\theta_p = \frac{1}{3} \langle (\overrightarrow{c_p} - \langle \overrightarrow{c_p} \rangle)^2 \rangle$ $\overrightarrow{q_{\theta_p}} = -k_{\theta_p} \nabla \theta_p$ $k_{\theta_p} = \frac{15(1 - \varepsilon_{l,b}) d_p \rho_p \sqrt{\pi \theta_p}}{4(41 - 33\eta)} \left[1 + \frac{12}{5} \eta^2 (4\eta - 3) (1 - \varepsilon_{l,b}) g_0 + \frac{16}{15\pi} (41 - 33\eta) \eta (1 - \varepsilon_{l,b}) g_0 \right]$ $\eta = \frac{1}{2} (1 + e_p)$

	$\gamma_{\theta_p} = \frac{12(1 - e_p^2)\rho_p g_0(1 - \varepsilon_{l,b})^2 \theta_p^{3/2}}{d_p \sqrt{\pi}}$
Characteristic Velocities	
Particle terminal velocity [75]	$U_t = \frac{U_*}{\left[\frac{\rho_w^2}{g\mu_w(\rho_p - \rho_w)} \right]^{1/3}}$ <p>where $U_* = \left(\frac{18}{d_*^2} + \frac{9.3393 - 6.9756\phi}{4d_*^{0.5}} \right)^{-1}$</p> $d_* = d_p \left[\frac{g\rho_w(\rho_p - \rho_w)}{\mu_w^2} \right]^{1/3}$
Minimum fluidization velocity [73]	$Re_{mf} = (1135.69 + 0.0408Ar)^{1/2} - 33.7$ <p>where $Re_{mf} = \frac{\rho_w U_{mf} d_p}{\mu_w}$ and $Ar = \frac{d_p^3 \rho_w (\rho_p - \rho_w)}{g\mu_w^2}$</p>
Boundary Condition	
Wall boundary condition [67]	$\overrightarrow{U_{p,s}} \cdot \overrightarrow{\bar{S}_p} \cdot \vec{n} + \frac{\sqrt{3}\pi}{6(1 - \varepsilon_{l,b})} \phi'(1 - \varepsilon)\rho_p g_{0,pp} \theta_p^{1/2} \overrightarrow{U_{p,s}} ^2 = 0$

Table 6.A2. Terminal velocities (U_t) of the particles for bidisperse GAC study in 3D AnFMBR.

Particle Diameter, d_p (mm)		Terminal Velocity, U_t [75] (m/s)	
d_{p1}	d_{p2}	U_{t1}	U_{t2}
0.9	1.1	0.0535	0.0631
0.8	1.2	0.0482	0.0675
0.7	1.3	0.0425	0.0717

Fig. 6.A1. Local amount of particle (n_p) with respect to reactor radius (r/R) at various binary GAC particle size ratio ($d_{p2}/d_{p1} = 1.22 - 1.86$) and axial positions ($h/H = 0.2 - 0.8$) for ratio of superficial water velocity to minimum fluidization velocity of binary mixture ($U_1/U_{mf,b}$) of (a) 2, (b) 6, and (c) 11.

Fig. 6.A2. Local particle momentum (p_p) with respect to reactor radius (r/R) for three bidisperse GAC mixtures ($d_{p2}/d_{p1} = 1.22, 1.50$, and 1.86) at three heights ($h/H = 0.2, 0.5$, and 0.8) for three superficial liquid velocity ratios ($U_l/U_{mf,b}$) of (a) 2, (b) 6, and (c)

CHAPTER 7: THE IMPACT OF POLYDISPERSE GAC FLUIDIZATION ON MEMBRANE FOULING MITIGATION IN ANFMBR

Abstract

The impact of GAC polydispersity types towards AnFMBR hydrodynamics and membrane fouling mitigation efficiency are investigated systematically in current study. Local hydrodynamics data from two-fluid model simulation on fluidization of three GAC mixtures (i.e., mono-, bi-, and tridisperse) with various polydispersity magnitude ($d_{p2}/d_{p1} = 1.18 - 1.78$ and $\sigma/d_{sm,m} = 0.1 - 0.5$) and mean particle diameters (1000 and 1400 μm) are correlated with fouling experiment results over wide range of inlet water velocities (0.0082 – 0.0385 m/s). Results show (i) GAC fouling mitigation effectiveness increases with mean particle diameter of polydisperse mixtures, while GAC-membrane scouring magnitude is also affected by the extend of GAC polydispersity, (ii) the number of GAC constituents (i.e., tridisperse > bidisperse > monodisperse) and size differences among GAC constituents (i.e., larger d_{p2}/d_{p1} and σ/d_{sm} for bidisperse and tridisperse, respectively) decrease membrane fouling mitigation efficiency and increase TMP for fully segregated condition (i.e., high inlet water velocity or power input), due to the higher pressure drop at dense bottom reactor occupied by larger GACs; the opposite trends of the were found in partially segregated condition (i.e., lower inlet water velocity or power input), presumably due to the higher inter-species collisions in such condition.

Keywords: anaerobic fluidized bed membrane bioreactor; polydispersity; two-fluid model; membrane fouling mitigation; hydrodynamics

7.1. Introduction

Recent studies had shown the benefits of fluidizing granular activated carbon (GAC) inside Anaerobic Fluidized bed Membrane Bio-Reactor (AnFMBR) as energy efficient membrane fouling mitigation process [1]. Micrometer-sized GAC prevents and reduces surface fouling mainly through scouring mechanism prior to GAC-membrane interactions, instead of GAC adsorption capability which is saturated over time. The effectiveness of membrane fouling mitigation depends on GAC properties and operating conditions, hence the need of correlating such variables and interspecies collisions and system hydrodynamics are guaranteed [2-4].

In comparison with the more common studied particle properties such as average particle size [5-8] and density [9, 10], there is a lack of detailed study on the impact of GAC polydispersity towards membrane fouling mitigation [11]. GAC manufactured as mixture of particles with different sizes (polydisperse form), but few studies had evaluated the impact of size polydispersity on GAC-membrane scouring inside AnFMBR or elaborated the polydispersity of GAC used within their studies. Subsequently, current study aims to systematically investigate the impact of size polydispersity by correlating hydrodynamics data (i.e., particle concentration/mixing, local particle velocity, and particle momentum) of mono-, bi-, and tridisperse GACs fluidization obtained from computational fluid dynamics (CFD) simulation, and fouling rate data (i.e., trans-membrane pressure / TMP) obtained from experiment. The impact of GAC polydispersity is investigated by (i) varying the number of GAC constituents (i.e., monodisperse, bidisperse, and ternary GACs) within similar size distribution range (i.e., $\sigma/d_{sm,m} = 0.1$ or 0.3) and average particle diameter (i.e., $1000\ \mu\text{m}$); and (ii) increasing PSD width of tridisperse mixtures (i.e., $\sigma/d_{sm,m} = 0.1 - 0.5$) at two Sauter-mean diameters (i.e., 1000 and $1400\ \mu\text{m}$).

7.2. Setup and Model

Figs. 7.1a and b showed the experimental and simulation set-up evaluated in current study. Complete experimental set-up has been described in our previous study [12] (see Fig. 7.1a) and it will be explained briefly here. 3D tubular AnFMBR column (d_c and h_c are 0.1 and 0.47 m, respectively) was operated with flat PVDF membrane (Merck Pte Ltd, nominal pore diameter = 0.22 mm, effective area = 0.0081 m²), model foulant solution of 2 g/L bentonite (Sigma-Aldrich Pte Ltd, mean particle diameter = 5.4 ± 0.11 μ m), and constant permeate flux of 30 L/m²/h (peristaltic pump Masterflex Cole-Parmer 7528-10, 6 – 600 rpm). Superficial water velocities (U_l) were adjusted and measured through Cole-Parmer's centrifugal pump and variable area flow-meter (1.5 – 20 L/min), respectively. Trans-membrane pressure (TMP) as an indicator of membrane fouling magnitude was measured with Cole-Parmer pressure transmitter (0 – 1 psi) and recorded on LabView (National Instrument Corp.) every 20 s. Reactor set-up similar to those of experimental study (Figs. 7.1a and b) was used in two-fluid model (TFM) simulation using open-source program of Multiphase Flow with interphase eXchange (MFiX) 2016-1 developed by National Energy Technology Laboratory (NETL) [13]. Complete simulation parameters and TFM equations are described in Table 7.1 and Appendix (Table 7.A1), respectively.

Fig. 7.1. Experimental (a) and CFD simulation (b) set-up of 3D AnFMBR.

Table 7.1. Simulation parameters used for polydisperse GAC study in 3D AnFMBR.

Parameter	Unit	Magnitude
Simulation time	s	80 (only the final 30 s is used for steady-state data analysis)
Time step	s	0.0001
Total number of grids	-	9600
Water Properties		
Water density (ρ_w)	kg/m ³	1000
Water viscosity (μ_w)	kg/m/s	0.001
Turbulent length scale (l)	m	0.01
Maximum turbulent viscosity (μ_{wt})	kg/m/s	100
GAC Properties		

GAC particle density (ρ_{pi} , similar for all particle constituents)	kg/m ³	1500
GAC particle constituent diameter (d_{pi})	m	(see Table 7.2)
Solid fraction above which viscous flow sets in ($\varepsilon_{p,min}$)	-	0.5
Solid fraction at maximum packing ($\varepsilon_{pi,max}$)	-	Bidisperse: Fedors-Landel equation [14] Tridisperse: Yu-Standish equation [15]
Initial slope of segregation (s_{coef} , for bidisperse and tridisperse particles) [16]	-	0.3
Particle-particle coefficient of friction (μ_k)	-	0.3
Particle-particle restitution coefficient (e_p)	-	0.99
Particle-wall restitution coefficient (e_r)	-	0.99
Angle of internal friction (ϕ)	°	30
Angle of internal friction at wall (ϕ_r)	°	11.31
Specularity coefficient at wall (ϕ') [17]	-	0.0001
Initial Conditions		
<i>Particle Bed</i>		
Bed height (h_0)	m	0.05
Void fraction ($\varepsilon_{l,b}$)	-	0.4
Solid fraction ($\varepsilon_{pi,b}$; $\varepsilon_{l,b} + \sum_{pi=1}^M \varepsilon_{pi,b} = 1$)	-	(see Table 7.2)
<i>Freeboard</i>		
Void fraction ($\varepsilon_{l,f}$)	-	1 (i.e., no GAC particles)

Boundary Conditions		
Bottom ($h/H = 0$): uniform superficial water velocity (U_l)	m/s	$8.2 \times 10^{-3} - 3.75 \times 10^{-2}$
Top ($h/H = 1$): uniform water pressure	Pa	101325
Sides ($r/R = -1$ and 1): partial slip for particle-wall interaction	-	Johnson-Jackson boundary conditions [18]

Subsequently, Table 7.2 lists details of GAC properties used in both experimental and simulation studies. Monodisperse, bidisperse, and tridisperse GAC mixtures with two average particle diameters (i.e., 1000 and 1400 μm), similar particle density (i.e., $\approx 1500 \text{ kg/m}^3$), and constant total bed mass (i.e., 350 g or $h_0 \approx 0.1 \text{ m}$) were fluidized at various inlet superficial water velocities (U_l). It is worth mentioning that the sizes and mass fractions of tridisperse mixture constituents are obtained by truncating mass-based frequency distribution of lognormal particle size distribution (PSD) function (see Fig. 7.2). The constituent diameter values of tridisperse mixtures are matched with available sieve tray opening to match experimental and simulation constituent diameters. U_l values used in current study are larger and smaller than minimum fluidization velocity (U_{mf}) and terminal velocity (U_t) of largest and smallest constituents, respectively. Such conditions allow fluidization of all particle constituents, while preventing entrainment or carry over of GAC constituents outside AnFMBR. Above mentioned set-up also enables evaluation of energy input (i.e., U_l values) towards AnFMBR performance, as energy loss due to bed pressure drop (i.e., similar total bed mass) and drag force (i.e., similar Sauter mean diameter) are constant.

Table 7.2. Detailed properties of mono-, bi-, and tridisperse GAC mixtures.

	Sieved GAC Diameter* (μm)	d_{pi} (μm)	$\varepsilon_{pi,b}$ (-)	U_{mf} Constituents (x 10 ⁻² m/s)	U_t Constituents (x 10 ⁻² m/s)
Monodisperse					
$d_p = 725 \text{ }\mu\text{m}$	(TFM)	725	0.6	0.15	4.39
$d_p = 1000 \text{ }\mu\text{m}$	(TFM)	1000	0.6	0.29	5.85
$d_p = 1290 \text{ }\mu\text{m}$	(TFM)	1290	0.6	0.45	7.13
Bidisperse					
$d_{p2}/d_{p1} = 1.18$	850 – 1000	925	0.3	0.21 – 0.39	5.09 – 6.67
	1000 – 1180	1090	0.3		
$d_{p2}/d_{p1} = 1.78$	600 – 850	725	0.3	0.11 – 0.52	3.63 – 7.56
	1180 – 1400	1290	0.3		
Tridisperse					
$\sigma/d_{sm,m}$ (1000 μm) = 0.1	850 – 1000	925	0.29	0.21 – 0.52	5.09 – 7.56
	1000 – 1180	1090	0.28		
	1180 – 1400	1290	0.03		
$\sigma/d_{sm,m}$ (1000 μm) = 0.3	500 – 850	675	0.16	0.07 – 0.72	2.97 – 8.64
	850 – 1180	1015	0.27		
	1180 – 1700	1440	0.18		
$\sigma/d_{sm,m}$ (1000 μm) = 0.5	425 – 1000	712.5	0.26	0.05 – 1.17	2.45 – 10.64
	1000 – 1400	1200	0.18		
	1400 – 2360	1880	0.17		
	710 – 1180	945	0.15	0.15 – 1.17	4.31 – 10.64

$\sigma/d_{sm,m}$ (1400 μm) = 0.3	1180 – 1700	1440	0.30		
	1700 – 2360	2030	0.16		
σ/d_{sm} (1400 μm) = 0.5	710 – 1400	1055	0.24	0.15 – 1.77	4.31 – 13.05
	1400 – 2000	1700	0.19		
	2000 – 3350	2675	0.17		

* Based on U.S.A standard sieves (ASTM specification E-11).

Fig. 7.2. Mass-based frequency of lognormal particle size distribution of GAC mixtures.

7.3. Results and Discussion

Fig. 7.3 compared expanded bed heights (i.e., total particle-membrane scouring area) from simulation and experimental studies of tridisperse mixtures, as current TFM codes have been validated for monodisperse and bidisperse mixtures in our previous studies [7]. Both experiment and simulation showed that expanded bed height increases with superficial water velocity (U_l), decreases with mean particle diameter ($d_{p,m}$), and relatively constant with polydispersity width. Higher U_l and smaller $d_{p,m}$ increases the relative impact of drag force ($U_l - U_{mf}$), hence increases expanded bed height. The

negligible impact of polydispersity width towards membrane scouring area (i.e., expanded bed height) warrants further study on interspecies interactions within expanded bed height, which affects the extent of local fouling mitigation. Simulation and experiment results also generally show good agreement, although differences among them increase with GAC mean diameter and PSD width. Such error could be explained through the size gaps among sieve trays, which increases with particle size, hence larger GAC constituents in experiment are less homogeneous in size.

Fig. 7.3. Comparison of expanded bed heights of experimental and simulation results.

7.3.1. Hydrodynamics of fluidized GAC mixtures

This section elaborates hydrodynamics data from TFM simulation which indicated the quality (i.e., GAC axial and lateral distribution, size segregation) and quantity (i.e., local GAC velocity, GAC momentum on membrane surface) of fouling mitigation process. Subsequently, Section 3.2. describes the correlation of such hydrodynamics data and membrane fouling rate (i.e., TMP) obtained from experimental results.

Table 7.3 displays the impact of number of GAC constituents (i.e., mono-, bi-, or tridisperse) for mixtures with similar mean diameter (i.e., 1000 μm), on expanded bed

height (h) and bulk density distribution (ρ_b) inside AnFMBR. Two observations highlighted here. First, despite the similarity of total expanded bed heights of mono-, bi-, and tridisperse mixtures for similar mean particle diameter, particle segregation in polydisperse mixtures affects local GAC concentration. Homogeneous axial and radial particle distribution in monodisperse GAC was found, compared to the general decreasing trend of GAC concentration for bidisperse and tridisperse mixtures axially. Specifically, larger constituents are segregated at lower column, and bed expansion of larger constituents is less than that of smaller constituents even for 50 % m/m bidisperse mixtures. Such results could be explained through the less prominent effect of water drag force on larger constituents ($U_l - U_{mf}$). This implies that smaller constituent in polydisperse mixture determine the area of GAC-membrane scouring (i.e., improve fouling mitigation qualitatively), while higher GAC concentration at lower column may increase GAC-membrane interactions and pressure drop compared to monodisperse system. Secondly, GAC constituents' expanded bed height and local particle concentration of fully segregated GAC mixtures (e.g., bidisperse $d_{p2}/d_{p1} = 1.78$) are similar to its constituents while fluidized separately (i.e., monodisperse $d_p = 725$ and $1290 \mu\text{m}$). On the other hand, fluidization behavior of GAC constituents in partially segregated system does not resemble those of the constituents fluidized separately (e.g., GAC constituent $d_p = 1015 \mu\text{m}$ in tridisperse $\sigma/d_{sm,m} = 0.3$, compared to monodisperse $d_p = 1000 \mu\text{m}$) due to inter-species interactions. The extent of particle segregation is affected by U_l (i.e., higher U_l tends to increase segregation), size difference among constituents diameter (i.e., larger d_{p2}/d_{p1} or $\sigma/d_{sm,m}$ increases segregation), and number of GAC constituents and constituents mass fraction (e.g., radial and axial segregation are observed for bidisperse $d_{p2}/d_{p1} = 1.18$ and tridisperse $\sigma/d_{sm,m} = 0.1$, respectively at

lower U_l of 0.0109 m/s, while the latter only differ in particle composition by 4.59 % m/m of 1290 μm). Overall this result displays the prominent impact of size polydispersity, even for mixtures with similar mean diameter and size distribution range, on AnFMBR hydrodynamics and particle-membrane scouring, but such factor is less investigated in previous studies.

Table 7.3. The impact of number of GAC constituents (i.e., mono-, bi-, or tridisperse) for GAC mixtures with $d_{p,m} \approx 1000 \mu\text{m}$ on expanded bed height (h) and contour plots of the bulk density (ρ_b).

Narrow Size Range	Monodisperse	Bidisperse $d_{p2}/d_{p1} = 1.18$		Tridisperse $\sigma/d_{sm,m} = 0.1$		
d_p (μm)	1000	925	1090	925	1090	1290
Mass (%)	100	50	50	49.16	46.25	4.59
$U_l = 0.0109 \text{ m/s}$
$U_l = 0.0245 \text{ m/s}$
Wide Size Range	Monodisperse	Bidisperse $d_{p2}/d_{p1} = 1.78$		Tridisperse $\sigma/d_{sm,m} = 0.3$

Similar to Table 7.3, Table 7.4 displays expanded bed height and bulk density distribution as function of PSD width (i.e., $\sigma/d_{sm,m} = 0.1 - 0.5$) and U_l (i.e., $0.01 - 0.0256$ m/s). Two results are mentioned here. Firstly, for similar PSD width, mean particle diameter of continuous PSD systems decreases expanded bed height but increases local particle concentration. Similar to previous result on monodisperse particle [7], larger mean diameter seems to increase membrane scouring at the expense of higher energy consumption. Secondly, while size difference among constituents diameters of bidisperse mixtures (d_{p2}/d_{p1} , see Table 7.3) increases axial gradient of particle concentration, axial segregation has plateau increasing trend with polydispersity width ($\sigma/d_{sm,m}$) of tridisperse mixtures at constant mean GAC diameter. Similar to previous

result for bidisperse mixture, membrane scouring area (i.e., total expanded bed height) does not vary with polydispersity of tridisperse mixture ($\sigma/d_{sm,m}$).

Table 7.4. The impact of PSD width (i.e., $\sigma/d_{sm,m} = 0.1 - 0.5$) on GAC expanded bed height (h) and contour plots of the bulk density (ρ_b).

	$\sigma/d_{sm,m}$ (1000 μm) = 0.1			$\sigma/d_{sm,m}$ (1000 μm) = 0.3			$\sigma/d_{sm,m}$ (1000 μm) = 0.5		
d_p (μm)	925	1090	1290	675	1015	1440	712.	1200	1880
Mass (%)	49.16	46.2	4.59	26.0	44.4	29.53	42.5	29.8	27.60
		5		2	5		7	3	
$U_l =$ 0.010 9 m/s
$U_l =$ 0.024 5 m/s
				$\sigma/d_{sm,m}$ (1400 μm) = 0.3			$\sigma/d_{sm,m}$ (1400 μm) = 0.5		
d_p (μm)				945	1440	2030	1055	1700	2675
Mass (%)				24.7	49.2	26.04	39.4	32.4	28.11
				4	3		0	9

The extent of GAC-membrane interactions does not only depend on the local GAC concentration, but also local GAC velocity. Fig. 7.4 elaborates local average particle velocity ($U_{p, ave}$) of mono-, bi-, and tridisperse mixtures with $d_{p,m} = 1000 \mu\text{m}$ and GAC size range within $\sigma/d_{sm,m} = 0.1$ versus dimensionless radial position (r/R) at three dimensionless axial positions (h/H) and superficial water velocities (U_l). $U_{p, ave}$ values of polydisperse mixtures (i.e., bidisperse and tridisperse) are obtained by mass-averaging local particle velocities of their constituents as obtained through TFM simulation. r/R represent ratio of radial position and radius of AnFMBR, while $r/R = 0$ and 1 indicate centre and wall AnFMBR, accordingly. Similarly, h/H represent ratio of axial position and total expanded bed height, while $h/H = 0$ and 1 indicate bottom and top expanded particle bed. Three conclusions are worth mentioning. Firstly, mono-, bi-, and tridisperse mixtures show relatively homogeneous $U_{p, ave}$ distribution radially, except at highest U_l value (0.0245 m/s) where GAC moves faster at centre and top column. Similarly, Table 7.3 showed that GACs tend to move towards centre column at highest U_l value (0.0245 m/s) presumably due to wall friction which decrease GAC velocity near wall. On the other hand, higher U_l caused complete axial segregation, hence larger constituents at lower column move slower than that of smaller constituents at top column due to the relative impact of drag force on them ($U_l - U_{mf}$). Secondly,

GAC polydispersity types affect $U_{p, ave}$ and such impact increases with U_l , due to the more prominent segregation phenomenon at higher U_l . It is worth reminding that complete segregation at high U_l implies $U_{p, ave}$ of polydisperse GAC at lower and upper column represent U_p of larger and smaller constituents, respectively. On the other hand, higher particle concentration at lower column for the higher number of GAC constituents (tridisperse > bidisperse > monodisperse) increases bed pressure drop, hence smaller GAC of polydisperse mixtures (925 μm) at top column have similar $U_{p, ave}$ value with those of monodisperse 1000 μm GAC. Thirdly, $U_{p, ave}$ of monodisperse GAC unsurprisingly increases with U_l , while the impact of U_l on $U_{p, ave}$ is less obvious for polydisperse mixtures. The latter could be explained from GAC local concentration (Table 7.3), specifically inter-species collisions and evolution of partially segregated system into complete segregation system as U_l increases.

Fig. 7.4. Local average particle velocity ($U_{p,ave}$) of narrow size distribution GAC mixtures of $d_{p,m} \approx 1000 \mu\text{m}$ ($d_p = 925 - 1290 \mu\text{m}$) with respect to reactor radius (r/R) at three heights ($h/H = 0.2, 0.5$ and 0.8) for three superficial water velocities (U_l) of (a) 0.0082 , (b) 0.0109 , and (c) 0.0245 m/s .

Similar to Fig. 7.4, Fig. 7.5 compares $U_{p,ave}$ distribution of mono-, bi-, and tridisperse mixtures for $d_{p,m} = 1000 \mu\text{m}$ and GAC size range within $\sigma/d_{sm,m} = 0.3$. Three observations worth discussing here. Firstly, expectedly, GAC diameter decreases $U_{p,ave}$ (i.e., $U_{p,ave}$ of monodisperse $1290 \mu\text{m} <$ that of $725 \mu\text{m}$) due to the decreasing relative impact of water drag ($U_l - U_{mf}$) on $U_{p,ave}$, and such effect is more pronounced at centre

column (i.e., no wall friction). Secondly, for all axial and radial positions, $U_{p,ave}$ of bidisperse $d_{p2}/d_{p1} = 1.78$ (which consist of 50 % m/m of 725 and 1290 μm GAC, see Table 7.2) is generally higher than $U_{p,ave}$ of monodisperse 725 μm GAC for partial segregation systems ($U_l = 0.0082$ and 0.0109 m/s), while bidisperse $d_{p2}/d_{p1} = 1.78$ shows generally lower $U_{p,ave}$ than that of monodisperse 1290 μm GAC for complete segregation system ($U_l = 0.0245$ m/s). The former presumably due to inter-species collisions within bidisperse GAC bed. The latter phenomenon caused by higher particle concentration of bidisperse mixture at lower column (area rich of larger constituents) which increases bed pressure drop, hence smaller constituents of bidisperse mixture (i.e., 725 μm) at upper column have lower $U_{p,ave}$ than that of monodisperse 1290 μm GAC. Thirdly, comparison between $U_{p,ave}$ of bidisperse $d_{p2}/d_{p1} = 1.18$ (Fig. 7.4) and 1.78 (Fig. 7.5) with similar $d_{p,m}$ shows that larger d_{p2}/d_{p1} mixture generally has larger and smaller $U_{p,ave}$ for partially ($U_l = 0.0082 - 0.0109$ m/s) and fully segregated systems ($U_l = 0.0245$ m/s), accordingly. The latter result on fully segregated condition is unexpected, because bidisperse $d_{p2}/d_{p1} = 1.78$ contains smaller GAC at upper column than that of $d_{p2}/d_{p1} = 1.18$ mixture, which implies larger $U_{p,ave}$ of bidisperse $d_{p2}/d_{p1} = 1.78$ at upper column. It is worth mentioning that bidisperse $d_{p2}/d_{p1} = 1.78$ has denser particle concentration at lower column for fully segregated condition ($U_l = 0.0245$ m/s) compared to that of $d_{p2}/d_{p1} = 1.18$, which caused higher pressure drop and decreases $U_{p,ave}$ of bidisperse $d_{p2}/d_{p1} = 1.78$ at upper column.

Fig. 7.5. Local average particle velocity ($U_{p,ave}$) of wide size distribution GAC mixtures of $d_{p,m} \approx 1000 \mu\text{m}$ ($d_p = 675 - 1440 \mu\text{m}$) with respect to reactor radius (r/R) at three heights ($h/H = 0.2, 0.5$ and 0.8) for three superficial water velocities (U_I) of (a) 0.0082 , (b) 0.0109 , and (c) 0.0245 m/s .

Fig. 7.6 further investigates the impact of polydispersity width ($\sigma/d_{sm,m}$) towards axial and radial $U_{p,ave}$ distribution of tridisperse mixtures with $d_{sm,m} = 1000$ and $1400 \mu\text{m}$ as superficial water velocity (U_I) increases. Similar to the results of bidisperse mixtures (Fig. 7.5), size differences among particle constituents (i.e., wider $\sigma/d_{sm,m}$) increases and decreases $U_{p,ave}$ for partial and complete segregation conditions, respectively. It is worth

highlighting that tridisperse $\sigma/d_{sm,m} (1000 \mu\text{m}) = 0.3 - 0.5$ and $\sigma/d_{sm,m} (1400 \mu\text{m}) = 0.3 - 0.5$ are under complete and partial segregation for $U_l = 0.0025 - 0.0026$ m/s (see Table 7.4), hence opposite impact of polydispersity width was found for these two GAC mixtures (Fig. 7.6c).

Fig. 7.6. Local average particle velocity ($U_{p,ave}$) of tridisperse GAC mixtures with different PSD widths ($\sigma/d_{sm,m} = 0.1 - 0.5$) and $d_{sm,m}$ (1000 – 1400 μm) with respect to reactor radius (r/R) at three heights ($h/H = 0.2, 0.5$ and 0.8) for three superficial water velocities (U_l) of (a) 0.0082, (b) 0.0109, and (c) 0.0245 m/s.

7.3.2. Relationship between fluidization hydrodynamics and fouling mitigation efficiency

Fluidized GAC scours membrane surface to prevent and reduce surface fouling formation from bentonite solution. Previous section has described GAC concentration and velocity distributions along reactor at various inlet liquid velocity, and the prominent impact of GAC polydispersity on system hydrodynamics. Current section correlated such hydrodynamics variables and membrane performance (i.e., trans-membrane pressure, or TMP). GAC-membrane interactions in current study is evaluated through total GAC momentum inside reactor (p) which is calculated as:

$$p = \sum_{pi=1}^M m_{pi} \cdot U_{pi,ave} \quad (1)$$

m_{pi} and $U_{pi,ave}$ represent particle mass (see Table 7.2) and average particle velocity (see Figs. 7.4 – 7.6) of GAC constituent pi inside AnFMBR, while M indicates number of GAC constituent (i.e., one for monodisperse, two for bidisperse, and three for tridisperse).

Subsequently, Fig. 7.7 plots total GAC momentum inside reactor (p) versus inlet water velocity (U_l) for various GAC polydispersity at narrow ($\sigma/d_{sm,m} \leq 0.1$, Fig. 7.7a) and wide PSD distributions ($\sigma/d_{sm,m} \leq 0.3$, Fig. 7.7b), and as PSD width increases ($\sigma/d_{sm,m} = 0.1 - 0.5$, Fig. 7.7c). Furthermore, total GAC momentum (p) is further correlated to fouling rate from experiment (i.e., $dTMP/dt$) in Fig. 7.7d. It is worth mentioning that limited inlet water velocity (< 0.013 m/s) due to pump limitation in experiment did not allow evaluation of fouling rate at higher U_l values. U_l values also represent pump energy supplied to the system, because energy loss due to bed pressure drop and drag force are constant at similar total bed mass and Sauter mean diameter conditions, respectively. Two conclusions from Fig. 7.7 are elaborated here. First, the impact of

polydispersity for similar mean GAC diameter is minimum at lower U_l (< 0.01 m/s) and it increases with U_l (Figs. 7.7a – c). Previous section on GAC local concentration (Tables 7.3 – 7.4) showed that GAC is partially segregated at lower U_l , hence GAC mean diameter seems to be sufficient to describe hydrodynamics characteristic of such systems. Similarly, the impact of GAC polydispersity is smaller for $d_{sm,m} = 1400$ μm than that of 1000 μm GAC because the former does not undergo complete segregation within U_l values evaluated here (see Table 7.4). Secondly, in agreement with previous results on the magnitude of local GAC concentration and velocity, total GAC momentum (p) generally decreases with number of constituents (tridisperse $<$ bidisperse $<$ monodisperse) and polydispersity magnitude (i.e., as d_{p2}/d_{p1} and σ/d_{sm} increase) for complete segregation condition (i.e., $U_l > 0.01$ m/s for $d_{sm,m} = 1000$ μm), and the opposite trends found for partial segregation condition. Fig. 7.7d further elaborates that higher p decreases fouling rate ($d\text{TMP}/dt$), hence alleviate surface fouling inside AnFMBR. This indicates that operation of AnFMBR using polydisperse GAC should avoid completely segregated condition to improve fouling mitigation.

Fig. 7.7. Comparison of overall particle momentum (p) versus superficial water velocity (U_l) of GAC mixtures with (a) narrow (925 – 1290 μm) and (b) wide (675 – 1440 μm) GAC size distribution; and (c) different polydispersity width ($\sigma/d_{sm,m}$).

7.4. Conclusion

Current study aims to systematically study the impact of GAC polydispersity on membrane fouling mitigation effectiveness inside AnFMBR. Specifically, GAC polydispersity was investigated by varying (i) the number of GAC constituents (i.e., mono-, bi-, and tridisperse), and (ii) the extent of GAC polydispersity (i.e., d_{p2}/d_{p1} and $\sigma/d_{sm,m}$ for bidisperse and tridisperse, respectively). Local hydrodynamics results (i.e., local particle concentration, particle velocity, and momentum) of fluidized monodisperse, bidisperse, and tridisperse GAC mixtures with various mean diameters

(1000 and 1400 μm) and polydispersities (i.e., $d_{p2}/d_{p1} = 1.18 - 1.78$ and $\sigma/d_{sm,m} = 0.1 - 0.5$) inside AnFMBR are correlated with membrane fouling rate data (i.e., TMP) from experimental results.

Two main conclusions are reiterated here. Firstly, mean diameter of polydisperse GAC decreases fouling rate, while the extend of GAC polydispersity does not affect membrane scouring area appreciably, but the effectiveness of particle-membrane scouring within expanded bed height. This underscores the importance of describing detailed particle polydispersity in future AnFMBR studies, as the effect of GAC polydispersity depends on both mean particle diameter and size range of particle constituents within the mixture. Secondly, the extend GAC polydispersity may affect particle-membrane scouring differently, depends on the extend of particle segregation. Specifically, the number of particle constituents (i.e., tri- > bi- > monodisperse) and polydispersity magnitude (i.e., d_{p2}/d_{p1} of bidisperse mixture, or $\sigma/d_{sm,m}$ of tridisperse mixture) reduce membrane fouling mitigation for completely segregated system (i.e., high inlet water velocity or power input), but the opposite effects were found in partially segregated system (i.e., lower inlet water velocity or power input). The former result caused by higher pressure drop at lower column (i.e., high particle concentration area) filled with larger GACs, while the latter result presumably due to the higher inter-species collisions as GAC constituents are partially mixed.

Nomenclature

A	= reactor cross section area, m^2
Ar	= Archimedes number, –
C_d	= particle drag coefficient, –
d^*	= dimensionless particle diameter, –
$d_{p,m}$	= mean particle diameter of mixture, mm
d_{p1}	= particle diameter of smaller constituent in bidisperse mixture, mm
d_{p2}	= particle diameter of larger constituent in bidisperse mixture, mm

D_c	= reactor diameter, m
e_p	= particle-particle restitution coefficient, -
e_r	= particle-wall restitution coefficient, -
g	= gravitational acceleration, m/s ²
g_0	= equilibrium radial distribution function at contact [19], -
h	= axial position in reactor, m
h_0	= initial particle bed height, m
H	= total expanded bed height, m
H_c	= reactor height, m
$J_{w,p}$	= granular temperature transfer between water and particle, kg/m/s ³
l	= turbulent length scale, m
M	= number of particle constituent in mixture excluding particle constituent s , -
$\vec{n}$	= unit normal vector from a boundary to particle, -
p_p	= local particle momentum, kg.m/s
$p_{w,p}$	= water-particle momentum transfer [13], kg.m/s
$q_{\theta p}$	= diffusive flux of granular temperature, kg/s ³
r	= radial position in reactor, m
P_w	= water pressure, Pa
R	= reactor radius, m
Re_{mf}	= Reynolds number at minimum fluidization, -
Re_p	= Reynolds number of particle, -
Re_w	= Reynolds number of water, -
U_*	= dimensionless particle velocity, -
U_l	= inlet superficial water velocity, m/s
U_{mf}	= minimum fluidization velocity, m/s
$\vec{U}_p$	= particle velocity vector, m/s
$U_{p,ave}$	= local average of particle velocity, m/s
U_{t1}	= terminal velocity of smaller particle, m/s
U_{t2}	= terminal velocity of larger particle, m/s
$\vec{U}_w$	= water velocity vector, m/s

Symbol and Greek Letters

β	= interphase momentum exchange coefficient, kg/m ³ /s
$\varepsilon_{l,b}$	= bed void fraction, -
$\varepsilon_{l,f}$	= freeboard void fraction, -
ε_p	= volume fraction of particle, -
μ_w	= water viscosity, kg/m/s
μ_{wt}	= maximum turbulent viscosity, kg/m/s
θ_p	= granular temperature, m ² /s ²
$\emptyset$	= angle of internal friction, °
$\emptyset_r$	= angle of internal friction at wall, °
$\emptyset'$	= specular coefficient at wall, -
ρ_w	= water density, kg/m ³
ρ_p	= particle density, kg/m ³
$\overline{\tau}_p$	= frictional-kinetic particle stress tensor [20], kg/m/s ²
$\overline{\tau}_w$	= water stress tensor [13], kg/m/s ²
$\gamma_{\theta p}$	= rate of dissipation of granular energy, kg/m/s ³

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Appendix

Table 7.A1. CFD equations used for polydisperse GAC study in 3D AnFMBR.

Continuity Equations	
Liquid phase	$\frac{\partial}{\partial t}(\varepsilon_w \rho_w) + \nabla \cdot (\varepsilon_w \rho_w \vec{U}_w) = 0$
Solid phase	$\frac{\partial}{\partial t}(\varepsilon_{pi} \rho_{pi}) + \nabla \cdot (\varepsilon_{pi} \rho_{pi} \vec{U}_{pi}) = 0$ <p>Where $pi = 1, 2, \dots, M$</p>
Momentum Equations	
Liquid phase	$\frac{\partial}{\partial t}(\varepsilon_w \rho_w \vec{U}_w) + \nabla \cdot (\varepsilon_w \rho_w \vec{U}_w \vec{U}_w)$ $= \nabla \cdot \vec{\tau}_w + \varepsilon_w \rho_w \vec{g} - \varepsilon_w \nabla P_w - \sum_{pi=1}^M \vec{I}_{w,pi}$
Solid phase	$\frac{\partial}{\partial t}(\varepsilon_{pi} \rho_{pi} \vec{U}_{pi}) + \nabla \cdot (\varepsilon_{pi} \rho_{pi} \vec{U}_{pi} \vec{U}_{pi})$ $= \nabla \cdot \vec{\tau}_{pi,pj} + \varepsilon_{pi} \rho_{pi} \vec{g} - \varepsilon_{pi} \nabla P_{pi} + \vec{I}_{w,pi} - \sum_{\substack{pj=1 \\ pj \neq pi}}^{M-1} \vec{I}_{pi,pj}$
Constitutive Equations	
Liquid-solid and solid-solid	<p>Between liquid and i^{th} solid phases:</p> $\vec{I}_{w,pi} = \beta_{w,pi}(\vec{U}_w - \vec{U}_{pi})$ <p>Between i^{th} and j^{th} solid phases:</p>

momentum exchange	$\vec{I}_{pi,pj} = \beta_{pi,pj}(\vec{U}_{pi} - \vec{U}_{pj})$ <p>where:</p> $\beta_{w,pi} = 18\mu_w(1 - \varepsilon_{pi})^2 \varepsilon_{pi} \frac{F}{d_{pi}^2}$ $F = 1 + \frac{3}{8} Re_{pi} \text{ , for } \varepsilon_{pi} \leq 0.01 \text{ and } Re_{pi} \leq \frac{(F_2 - 1)}{(3/8 - F_3)}$ $F = F_0 + F_1 Re_{pi}^2 \text{ , for } \varepsilon_{pi} > 0.01 \text{ and } Re_{pi} \leq \frac{F_3 + \sqrt{F_3^2 + 4F_1(F_0 - F_2)}}{2F_1}$ $F = F_2 + F_3 Re_{pi} \text{ , for } \begin{cases} \varepsilon_{pi} \leq 0.01 \text{ and } Re_{pi} > \frac{(F_2 - 1)}{(3/8 - F_3)} \\ \varepsilon_{pi} > 0.01 \text{ and } Re_{pi} > \frac{F_3 + \sqrt{F_3^2 + 4F_1(F_0 - F_2)}}{2F_1} \end{cases}$ <p>F_1, F_2, and F_3 are drag force coefficients [13].</p> $\beta_{pi,pj} = 3(1 + e_p) \left(\frac{\pi}{2} + \mu_k \frac{\pi^2}{8} \right) \frac{(d_{pj} + d_{pi})^2}{2\pi(\rho_{pj}d_{pj}^3 + \rho_{pi}d_{pi}^3)} \rho_{pi}^2 g_0 \vec{U}_{pj} - \vec{U}_{pi} + s_{coef} P_c$ $P_c \begin{cases} 10^{24}(\varepsilon_{l,mf} - \varepsilon_w)^{10} \text{ for } \varepsilon_w < \varepsilon_{l,mf} \\ 0.05 \frac{(\varepsilon_{pi} - \varepsilon_{p,min})^2}{(\varepsilon_w - \varepsilon_{l,mf})^5} \text{ for } \varepsilon_{l,mf} \leq \varepsilon_w < (1 - \varepsilon_{p,min}) \\ 0 \text{ for } \varepsilon_w \geq (1 - \varepsilon_{p,min}) \end{cases}$
Granular temperature [21]	$\frac{3}{2} \left[\frac{\partial}{\partial t} (\varepsilon_{pi} \rho_{pi} \theta_{pi}) + \nabla \cdot (\varepsilon_{pi} \rho_{pi} \theta_{pi} \vec{U}_{pi}) \right]$ $= \left[\overline{\overline{\tau}}_{pi} : \nabla \vec{U}_{pi} - \nabla \cdot \vec{q}_{\theta_{pi}} - \gamma_{\theta_{pi}} + J_{w,pi} \right]$ <p>where</p> $\theta_{pi} = \frac{1}{3} \langle (\vec{U}_{pi} - \langle \vec{U}_{pi} \rangle)^2 \rangle$

	$\overrightarrow{q_{\theta_{pi}}} = -k_{\theta_{pi}} \nabla \theta_{pi}$ $k_{\theta_{pi}} = \frac{15\varepsilon_{pi}d_{pi}\rho_{pi}\sqrt{\pi\theta_{pi}}}{4(41-33\eta)} \left[1 + \frac{12}{5}\eta^2(4\eta-3)\varepsilon_{pi}g_0 \right. \\ \left. + \frac{16}{15\pi}(41-33\eta)\eta\varepsilon_{pi}g_0 \right]$ $\eta = \frac{1}{2}(1+e_p)$ $\gamma_{\theta_{pi}} = \frac{12(1-e_p^2)\rho_{pi}g_0\varepsilon_{pi}^2\theta_{pi}^{3/2}}{d_{pi}\sqrt{\pi}}$
Boundary Condition	
Wall boundary condition [18]	$\overrightarrow{U_{pi}} \cdot \overrightarrow{\tau_{pi}} \cdot \vec{n} + \frac{\sqrt{3}\pi}{6\varepsilon_{pi}} \theta'_{\varepsilon_{pi}\rho_{pi}g_0\theta_{pi}^{1/2}} \overrightarrow{U_{pi}} ^2 = 0$

CHAPTER 8: CONCLUSIONS AND FUTURE PERSPECTIVES

8.1. Conclusions

Despite the broad impacts of particle polydispersity on fluidization phenomena, systematic study on such variable is overshadowed by the more common particle properties such as mean particle size and density of the mixture. The complex and chaotic nature of fluid-solid interactions within fluidized bed systems further hinder such effort, resulted in contradictory findings on the impact of particle polydispersity and the abundant amount of semi empirical design correlations which require in-field adjustment upon its application. Mechanistic understanding on the impact of particle polydispersity towards fluidization phenomena is guaranteed to improve the design and operation of current and future fluidized bed systems. This thesis aims to systematically evaluated the impact of particle polydispersity (i.e., monodisperse, binary size/density, ternary mixture, and continuous PSD) on flow hydrodynamics (i.e., local particle and fluid velocities, particle mixing/segregation, inter-species collisions), key fluidization phenomena (i.e., entrainment, TDH, particle clustering), and fluidized bed performance (i.e., energy consumption, membrane fouling mitigation in fluidized bed membrane bioreactor).

8.1.1. Particle polydispersity effects on current predictive capability of fluidization phenomena (entrainment flux, TDH, particle clustering)

Chapters 2 – 4 focus on the impact of particle polydispersity on key phenomena affecting fluidized bed design, namely particle entrainment, transport disengaging height (TDH), and cluster characteristics. Evaluation of entrainment flux and TDH values of monodisperse, binary, and continuous PSD mixtures indicate (i) the use of

empirical constants and mean size or density of particle mixtures are inadequate to represent polydispersity impact on fluidization phenomena, as demonstrated by huge discrepancies among prediction of design correlations developed from similar set ups; (ii) particle polydispersity affects fluidization behaviour through inter-species interactions, namely cohesive agglomeration of fine constituents, particle clustering in the presence of hydrodynamics instability, and inter-species collisions, which guarantee systematic studies on such topics.

Comprehensive evaluation on cluster characteristics of various particle polydispersities in circulating fluidized bed (CFB) riser found (i) within the well mixed area, the more dominant inter-species collisions in polydisperse systems seems to promote cluster break-up more than that of monodisperse system; (ii) size segregation of polydisperse mixtures (i.e., smaller and larger constituents at centre and near wall column) hinders cluster formation (i.e., cluster formation of binary < continuous PSD), as larger constituents showed lower cluster appearance probability compared to smaller constituents, while smaller-constituents clusters are breaking up as it travels to wall area (i.e., area rich of larger constituents).

8.1.2. CFD study on the impact of particle polydispersity on fluidized bed system performance.

Chapters 5 – 7 evaluate fluidization behavior of monodisperse, bidisperse, and tridisperse granular activated carbon (GAC) and its correlations with particle-membrane scouring efficiency as fouling mitigation mechanism in Anaerobic Fluidized Bed Membrane Bioreactor (AnFMBR). Computational Fluid Dynamics (CFD) results on system hydrodynamics and fouling experiment data are used to correlate inter-species interactions of mono- and polydisperse systems, and AnFMBR performance (i.e., trans-

membrane pressure (TMP) and energy consumption). Three main conclusions worth highlighting here, (i) larger GAC mean diameter increases particle-membrane momentum transfer (i.e., membrane scouring) due to the larger particle inertia, and the higher superficial liquid velocity required to cover entire membrane height, which indicate the trade-off between fouling effectiveness and energy consumption in AnFMBR; (ii) in fully segregated system (i.e., high superficial liquid velocity or power input), the higher number of particle constituents of GAC mixtures (i.e., tridisperse > bidisperse > monodisperse) and the wider size differences among particle constituents (i.e., larger d_{p2}/d_{p1} and σ/d_{sm} for bidisperse and continuous PSD, respectively) decrease membrane scouring effectiveness (or increase TMP), because smaller constituents at top reactor received less energy due to higher pressure drop at bottom reactor occupied by highly-concentrated larger constituents; (iii) in partially mixed system (i.e., lower superficial liquid velocity), the opposite trends of the (ii) conclusion were tended to be found, presumably due to higher inter-species collisions. Regarding conclusions number (ii) and (iii), two concealed messages are worth highlighting. Firstly, conclusions number (ii) and (iii) indicate the prominent impact of particle polydispersity on fluidized bed system performance, and such impact depends on the extent of particle segregation within the system. This also implies that the choice of operating conditions (i.e., U_l) in AnFMBR does not only depend on the evaluation of pump energy, but also the segregation of polydisperse mixture inside AnFMBR. Current study showed that polyidpserse GAC reduces less fouling than its monodisperse counterpart when the former is fully segregated. Secondly, particle polydispersity showed opposite impacts on fluidization system performance as operating conditions and particle properties changes, which underscored the importance of systematic study and comprehensive data

mining to cultivate mechanistic understanding on the impact of particle polydispersity, and to avoid contradictory results among different studies (i.e., Chapters 2 – 3).

8.2. Future Perspectives

Chapters 2 and 3 evaluate predictive capabilities of available entrainment and TDH correlations using various particle polydispersity types. Such versatility is desired to improve cyclone and fluidized bed designs and minimize the lost of valuable catalyst particles in industry. These chapters unveiled (i) the need to incorporate particle polydispersity into fluidization correlations to improve its versatility, beyond the use of semi empirical constants and mean-particle diameter and/or density of the mixtures; (ii) systematic study on the relation between inter-species interactions of polydisperse mixture's constituents and mesoscale phenomena (e.g., cohesive agglomerates, clusters) could improve mechanistic understanding on the impact of polydispersity towards macro behavior of fluidized bed.

Chapter 4 investigates the relationship between bed polydispersity and cluster characteristics in CFB riser. This chapter underscored (i) comprehensive study (e.g., local sampling at various radial and axial positions, detailed report on the polydispersity characteristics of particle bed) and standardized cluster measurement methods (e.g., minimum threshold of cluster detection) are needed to avoid contradictory finding among studies on cluster characteristics, despite the interplay of a host of factors on such mesoscale phenomenon; (ii) systematic study on the impact of shape polydispersity (e.g., variation of particle sphericity/other shape factor) towards cluster characteristics is needed, while there are few comprehensive data available for the impacts of size polydispersity (e.g., variation of size or density ratio of binary mixture, polydispersity width of continuous PSD).

Chapters 5 – 7 elaborate CFD study on the impact of fluidizing various polydispersity types of GAC (i.e., mono-, bi-, and tridisperse mixtures) towards AnFMBR hydrodynamics, particle-membrane scouring, and membrane fouling mitigation. In view of relatively new development of AnFMBR system, and the very few study of simulation work in membrane field, a lot of opportunities are available for future studies both in experimental and simulation studies. Few of such recommendations are mentioned as follow, (i) the incorporation of baffles inside AnFMBR to aid particle mixing of polydisperse system and system hydrodynamics, (ii) a more detailed study relating inter-species collisions and particle-membrane collisions inside AnFMBR, which could not be obtained from TFM codes, but easily evaluated from CFD-DEM code; (iii) the use of more complex liquid physical properties in simulation study to mimic wastewater stream (e.g., higher viscosity/density, the presence of particulate foulants or oil emulsion); (iv) simulation study using hollow fibre membranes in which dynamic fibre movement affecting both system hydrodynamics and particle-membrane-interactions.

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