



Titre: Quantification of the Magnitude of Interparticle Forces in a Gas-Solid
Title: Fluidized Bed

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Date: 2023

Type: Mémoire ou thèse / Dissertation or Thesis

Référence: Soleimani, I. (2023). Quantification of the Magnitude of Interparticle Forces in a
Citation: Gas-Solid Fluidized Bed [Thèse de doctorat, Polytechnique Montréal]. PolyPublie.
<https://publications.polymtl.ca/55722/>

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Programme: Génie chimique
Program:

POLYTECHNIQUE MONTRÉAL

affiliée à l'Université de Montréal

**Quantification of the Magnitude of Interparticle Forces in a Gas-Solid
Fluidized Bed**

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Thèse présentée en vue de l'obtention du diplôme de *Philosophiae Doctor*

Génie chimique

Août 2023

POLYTECHNIQUE MONTRÉAL

affiliée à l'Université de Montréal

Cette thèse intitulée :

Quantification of the Magnitude of Interparticle Forces in a Gas-Solid Fluidized Bed

présentée par **Iman SOLEIMANI**

en vue de l'obtention du diplôme de *Philosophiae Doctor*

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DEDICATION

To my beloved ones.

ACKNOWLEDGEMENTS

I would like to extend my gratitude to everyone who contributed to the successful completion of this project.

I want to express my sincere appreciation to my supervisors, Prof. Jamal Chaouki, and Dr. Jaber Shabanian, for their invaluable guidance, encouragement, and patience throughout my challenging Ph.D. journey. Working with them has been an honor and a tremendous learning experience, beyond what is included in this thesis.

I would also like to thank Prof. Gregory S. Patience, Prof. Louis Fradette, and Dr. Raymond A. Cocco, for accepting to serve as members of the thesis committee.

I am sincerely grateful to my dear friend, Majid Rasouli, for his incredible support even before my journey began. He introduced me to my supervisors and provided unwavering support during my early days in Montreal. Thank you, Majid, for being such an invaluable friend.

I would like to extend my gratitude to all members of our research group (PEARL), in particular, Dr. Mohammad Latifi, Dr. Mojtaba Mokhtari, Dr. El Mahdi Lakhdisi, Dr. Hamed Nasri, and Dr. Rouzbeh Jafari for valuable discussions, and Mr. Ahmed Najjar for helping to translate the abstract into French. Special thanks to the technicians of the Chemical Engineering department, particularly Robert Delisle, Sylvain Fleury-Simard, and Maxime Beaudoin. Their technical expertise and support have been instrumental in the success of this project.

To my family: your love, understanding, and sacrifices have been crucial in my academic success, and I am forever grateful for your presence in my life. I want to express my deep gratitude to my siblings for their selfless actions in fulfilling my responsibilities when I couldn't be there. I am grateful to my mother, who played a vital role as my first teacher and has been a constant source of motivation and inspiration since the very beginning of my life.

Last but certainly not least, I want to express my deepest appreciation to my girlfriend for her invaluable support throughout this journey. Her selfless dedication has been truly priceless. I cannot find adequate words to express my feelings of gratitude for you, my beloved Saba.

RÉSUMÉ

Les réacteurs à lit fluidisé de type gaz-solide sont largement utilisés dans de nombreuses industries chimiques en raison de leurs excellentes performances en termes de transfert de masse et de chaleur, dépassant ainsi celles d'autres contacteurs gaz-solide. Ces performances sont étroitement liées à l'hydrodynamique du lit, elle-même déterminée par l'intensité des forces inter particules (IPFs) et des forces hydrodynamiques (HDFs). Ces forces exercent une influence sur la conception, la mise à l'échelle, le fonctionnement et les performances globales du lit. Les IPFs exercent un impact significatif sur les caractéristiques hydrodynamiques du lit dans diverses situations, particulièrement dans les réacteurs à lit fluidisé catalytique. Ces situations comprennent notamment le groupe A de la classification de Geldart, où les IPFs sont du même ordre de grandeur que les HDFs. De même, elles incluent le groupe C de la classification de Geldart, où les IPFs contrôlent le comportement hydrodynamique, et le fonctionnement à haute température, où l'ampleur des IPFs peut être beaucoup plus importante que dans les conditions ambiantes. Les IPFs et leurs effets sur l'hydrodynamique du réacteur à lit fluidisé ne sont pas toujours bien étudiés, en raison du manque de techniques de mesure permettant une quantification adéquate de ces forces dans le cas d'un lit fluidisé de type gaz-solide.

En conséquence, cette étude se concentre principalement sur le développement d'approches fiables visant à quantifier avec précision l'ampleur des IPFs dans un lit fluidisé type gaz-solide. Par la suite, une intégration de l'effet des IPFs dans la classification de Geldart est envisagée afin de caractériser de manière rigoureuse la frontière entre les groupes A et C de la classification de Geldart.

Trois approches ont été introduites pour quantifier l'ordre de grandeur des IPFs dans un lit fluidisé gaz-solide. La première approche est basée sur l'augmentation de la vitesse minimale expérimentale de fluidisation $U_{mf,exp}$ avec les IPFs. $U_{mf,exp}$ s'écarte donc de la prédiction d'une équation purement hydrodynamique $U_{mf,pred}$. Cette dernière est estimée en égalisant le poids du lit et la force de traînée exercée par le flux de gaz. La différence entre $U_{mf,exp}$ et $U_{mf,pred}$ est par conséquent le débit de gaz nécessaire pour surmonter les IPFs. Par la suite, il a été démontré que

le rapport entre les IPF agissant sur une particule et son poids W_p était en corrélation avec le ratio entre $U_{mf,exp}$ et $U_{mf,pred}$. Cette relation est appelée l'équation de "l'écart U_{mf} ".

La deuxième approche repose sur l'apparition d'un dépassement dans le profil de la chute de pression du lit à la fluidisation minimale, qui est attribué à la différence entre le vide en vrac lâche ε_0 et le vide de fluidisation minimale ε_{mf} . Le ratio $\varepsilon_{mf}/\varepsilon_0$, appelé ratio dynamique de Hausner (DHR), a été démontré comme représentant les IPFs qui sont surmontées pendant la fluidisation. En conséquence, le rapport $IPFs/W_p$ a été corrélé avec le rapport $\varepsilon_{mf}/\varepsilon_0$ dans l'équation DHR. Cette équation sous-estime l'ampleur des IPFs dans la fluidisation des particules agglomérées. La forme généralisée de l'équation DHR a ensuite été élaborée en considérant le ratio des diamètres des agglomérats et des particules comme représentatif de la fraction des IPFs qui agissent encore sur les particules après la fluidisation. Les résultats montrent que les IPFs peuvent être supérieurs à W_p par quatre ordres de grandeur pour les poudres ayant un comportement du groupe C de Geldart et sont du même ordre de grandeur si le comportement de fluidisation du groupe A de Geldart est observé.

La troisième approche repose sur la variation du vide du lit engendrée par la formation d'agglomérats résultant de la présence de IPFs. Le modèle proposé présente une version modifiée du ratio de Hausner (MHR) en tant que paramètre essentiel pour établir des relations quantitatives entre le vide du lit, la taille des agglomérats et l'ordre de grandeur des IPFs. Dans le cas des poudres appartenant au groupe C de la classification de Geldart, le MHR correspond au rapport entre la densité des particules et la densité taraulée. Les résultats obtenus à l'aide du modèle MHR démontrent l'existence de deux équations : l'une établit une relation proportionnelle entre le rapport des diamètres des agglomérats et des particules, et le carré du MHR ; tandis que l'autre montre que le rapport des $IPFs/W_p$ varie avec MHR à la puissance quatre.

En utilisant les équations proposées pour quantifier les IPFs, un nouveau modèle a été proposé pour caractériser la frontière entre les comportements des groupes A et C de la classification de Geldart. Le critère de choix suppose que la vitesse du gaz interstitiel doit dépasser la vitesse terminale de chute des particules pour passer du comportement du groupe A au comportement du groupe C de la classification de Geldart. La combinaison de cette hypothèse avec les équations de déviation généralisées du DHR et U_{mf} a conduit à deux équations équivalentes. Elles expriment l'ordre de

grandeur des IPFs et le rapport des diamètres des agglomérats et des particules à la frontière entre les deux comportements caractéristiques en fonction de ε_{mf} . Les données expérimentales provenant de la littérature ont montré que le comportement de fluidisation bascule entre les groupes A et C de la classification de Geldart lorsque les IPFs sont d'un ordre de grandeur supérieur à W_p .

ABSTRACT

Many chemical industries take advantage of gas-solid fluidized beds because of their superior heat and mass transfer characteristics compared to other gas-solid contactors. These characteristics are highly impacted by bed hydrodynamics, which, in turn, determined by the magnitudes of interparticle forces (IPFs) and hydrodynamic forces (HDFs). These forces and their relative importance ultimately affect the bed design, scale-up, operation, and overall performance. IPFs significantly affect the bed hydrodynamics in several situations, which are common in catalytic and non-catalytic gas-solid fluidized bed reactors. They include Geldart group A fluidization, where IPFs are in the same order of magnitude as HDFs, Geldart group C fluidization, where IPFs govern hydrodynamic behaviour, and high-temperature operation, where the magnitude of IPFs can significantly be greater than that at ambient conditions. IPFs and their influence on the fluidized bed hydrodynamics are not completely understood since no measurement technique was available to adequately quantify these forces in a gas-solid fluidized bed.

This study focuses on developing reliable and non-disrupting approaches for quantifying the magnitude of IPFs in a gas-solid fluidized bed. Incorporating the effect of IPFs into Geldart classification to characterize the boundary between Geldart groups A and C is attempted in the second part of this study.

Three approaches were introduced to quantify the magnitude of IPFs in a gas-solid fluidized bed. The first is based on the increase of the experimental minimum fluidization velocity $U_{mf,exp}$ with IPFs. $U_{mf,exp}$, consequently, deviates from the prediction of a purely hydrodynamic equation $U_{mf,pred}$. The latter is estimated by equating the weight of the bed and the drag force exerted by the gas flow. The difference between $U_{mf,exp}$ and $U_{mf,pred}$, therefore, is the gas flow required to overcome IPFs. The ratio of IPFs acting on a particle and its weight W_p was ultimately shown to be correlated with the ratio of $U_{mf,exp}$ and $U_{mf,pred}$. The relationship was called the " U_{mf} deviation" equation.

The second approach was established on the appearance of an overshoot in the bed pressure drop profile at minimum fluidization, which was attributed to the difference between the loose bulk

voidage ε_0 and minimum fluidization voidage ε_{mf} . The ratio of $\varepsilon_{mf}/\varepsilon_0$, which was called the dynamic Hausner ratio (DHR), was shown to represent IPFs that are overcome during fluidization. Accordingly, the ratio of $IPFs/W_p$ was correlated with $\varepsilon_{mf}/\varepsilon_0$ in the DHR equation. This equation underestimated the magnitude of IPFs in agglomerated particles fluidization. The generalized form of the DHR equation was subsequently developed considering the ratio of agglomerate and particle diameters as a representative of the fraction of IPFs that remain acting on particles after fluidization. Results indicated that IPFs can be greater than W_p by four orders of magnitude for powders with Geldart group C behaviour and are in the same order of magnitude if the fluidization behaviour of Geldart group A is observed.

The third approach was founded on the variation of the bed voidage due to the formation of agglomerates because of the presence of IPFs. The presented model introduces a modified version of the Hausner ratio (MHR) as a crucial parameter for establishing quantitative relationships between bed voidage, agglomerate size, and the magnitude of IPFs. In the case of Geldart group C powders, the MHR is equal to the ratio of particle density to the tapped bulk density. The results of the MHR model are two equations demonstrating that the ratio of agglomerate and particle diameters is proportional to the square of the MHR, while the ratio of $IPFs/W_p$ varies with the fourth power of the MHR.

Employing the proposed equations for quantifying IPFs, a new model was proposed for characterizing the boundary between Geldart groups A and C. The criterion assumes that the interstitial gas velocity must exceed the particle terminal velocity for the transition from the Geldart group A to C behaviour. Combining this assumption with the generalized DHR and U_{mf} deviation equations led to two equivalent equations. They express the magnitude of IPFs and the ratio of agglomerate and particle diameters at the boundary between the two characteristic behaviours as a function of ε_{mf} . The experimental data from literature showed that the fluidization behaviour is switched between Geldart groups A and C when IPFs are an order of magnitude greater than W_p .

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CHAPTER 1 INTRODUCTION

Gas-solid fluidized beds are among the original technologies for processing particulate materials. These units are broadly employed in chemical industries, including fluid catalytic cracking (FCC), gas-phase polymerization, solid combustion, and gasification, as well as emerging technologies such as oxy-fuel combustion, chemical looping, and CO₂ hydrogenation to methanol [1, 2].

Fluidization is essentially the process of suspending solid particles in a gas flow and transforming the particulate material into a fluid-like state where particles can move. The fluidized solid materials demonstrate beneficial features, including easy handling and the capability of solid flow between vessels, enhanced heat and mass transfer, and lower pressure drop at a given gas velocity [1, 2]. The heat and mass transfer coefficients in a gas-solid fluidized bed are substantially greater than those in other gas-solid contactors [2, 3]. This is a key advantage of gas-solid fluidized beds leading to an increased overall process rate and a uniform and controllable temperature and, ultimately, to the wide application of these units in solid processing and catalytic gas-phase reactions. These favorable characteristics are, however, significantly influenced by the bed hydrodynamics, i.e., the gas flow pattern and solid motion in the bed. Therefore, a comprehensive understanding of the hydrodynamic behaviour of particulate materials in gas-solid fluidized beds is essential for optimal process design, improving product quality, and driving innovation in the corresponding industries.

The dynamics of particles and, consequently, hydrodynamics of a gas-solid fluidized bed is determined by all forces acting on the particles and their relative magnitudes [4-6]. These forces include gravitational force, hydrodynamic forces (HDFs), and interparticle forces (IPFs) [6, 7]. HDFs originate from gas-solid interaction and include buoyancy and drag forces as the countervailing force to overcome gravity [6, 7]. Hence, gravitational and HDFs are of the same order of magnitude in the fluidized state of the bed. This is manifested in the similarity of the gas pressure drop at the fluidized state and the equivalent pressure drop, which is determined by dividing the weight of the bed material by its cross-sectional area. Accordingly, the determining factor for the hydrodynamic behaviour of the bed is the magnitude of IPFs compared to gravitational or HDFs [4, 5]. For instance, it was suggested that for naturally cohesive particles

with Geldart [8] group C behaviour, the magnitude of IPFs experienced by a particle is substantially greater than the gravitational force [4, 5, 9]. Hence, IPFs dominate the fluidization behaviour of these powders. In contrast, IPFs are unimportant compared to gravitational or HDFs acting on particles with Geldart group B behaviour and become practically negligible for large particles with Geldart group D behaviour [4, 5]. The hydrodynamic behaviours of these particles are, hence, governed by gravitational and HDFs. Particles with the typical behaviour of Geldart group A fall between the above two extremes where IPFs, HDFs, and gravity are in the same order of magnitude [4-6].

IPFs can significantly influence the hydrodynamics of a gas-solid fluidized bed in several circumstances. A common example is the fluidization of fine and ultrafine powders with Geldart groups A or C behaviours. These particles are typically employed in gas-solid fluidized beds for catalytic reactions owing to their high specific surface area and favorable fluidization behaviour [1, 10]. Operating conditions can also contribute to the magnitude of IPFs [5, 11]. For example, an increase in the operating temperature can amplify van der Waals forces [11-14]. In addition, the formation of liquid/solid bridges at temperatures close to the minimum sintering temperature of particles leads to substantial growth in the total magnitude of IPFs [5, 15]. Consequently, a particulate assembly with the typical behaviour of Geldart group B or D at ambient conditions can show group A or C characteristics at high temperatures [5, 14, 16, 17]. Therefore, it is crucial to consider the variation of both IPFs and HDFs for a reliable prediction of the bed hydrodynamic behaviour at elevated operating conditions.

Despite the crucial importance of IPFs, they have been largely neglected for the prediction of the hydrodynamics of gas-solid fluidized beds. Neglecting the effect of IPFs is the fundamental reason behind the extensive inconsistency in the experimental results on the impact of operating conditions on the bed hydrodynamics [5]. Application of purely hydrodynamic correlations in cases where the magnitude of IPFs is not negligible leads to significant deviations from experimental data [18-21]. Geldart classification also becomes invalid if the magnitude of IPFs changes significantly [22]. The design, scale-up, and operation of gas-solid fluidized beds are highly impacted by the Geldart behaviour of the bed and its hydrodynamic parameters [1, 2, 10]. Hence, inaccurate prediction of the bed hydrodynamic parameters and its Geldart behaviour caused by neglecting or unreliable

estimation of IPFs can significantly affect the process and may lead to unit shutdown under extreme conditions [23].

The lack of an accurate measurement method to quantify the effective IPFs between particles in a gas-solid fluidized bed is the primary cause of the limited knowledge of IPFs and their influences on fluidized bed hydrodynamics. Although several experimental methods and theoretical models have been developed for quantitative and qualitative evaluation of IPFs, they are hardly applicable to the conditions of an aerated particulate assembly in a gas-solid fluidized bed. Consequently, applying these methods to quantify IPFs for particles close to the boundaries between Geldart groups C, A, and B led to inconsistent and unrealistic results. For instance, the proposed ratios of IPFs and weight of a particle at the boundary of Geldart groups A and C differ by orders of magnitude, i.e., from 19 [16] to about 5800 [11]. The significance of in-situ approaches for quantification of IPFs in a gas-solid fluidized bed is highlighted by the importance of accurate hydrodynamic correlations for an appropriate design and scale-up on the one hand and the inconsistent results produced by conventional IPFs measurement methods on the other.

Therefore, this study aims to develop fundamental knowledge for the quantitative investigation of IPFs in gas-solid fluidized beds, their variation with operating conditions, and their effects on the bed hydrodynamics. Hence, the primary objective of this thesis is to develop reliable in-situ approaches for quantifying IPFs in a gas-solid fluidized bed. These approaches will serve as the basis for developing reliable hydrodynamic correlations and research on the Geldart behaviour under varying magnitudes of IPFs.

CHAPTER 2 LITERATURE REVIEW

Gas-solid fluidization is the process of transforming a particulate assembly into a fluid-like material where particles can move like molecules of a liquid. This is achieved by suspending solid particles in a gas flow. Thus, fluidization is the state of balance between forces exerted on particles. These include hydrodynamic forces (HDFs), gravitational, and interparticle forces (IPFs) [1, 2]. This chapter is dedicated to these forces, particularly IPFs, the effects of IPFs on bed hydrodynamics, and the available approaches for measuring these forces.

2.1 Hydrodynamic forces

HDFs originate from solid-fluid interaction and have several types that may be important under specific conditions. Lift forces, for instance, act normal to the flow direction and are either due to the rotation of a particle or velocity gradient in the fluid flow [3, 4]. Important HDFs in fluidization are the drag and buoyancy forces.

2.1.1 Drag force

Drag force acts in the direction of the slip velocity [5], i.e., the difference between particle and fluid velocities. The drag force on a stagnant particle with a diameter d_p in a uniform flow field is expressed by [2]:

$$F_D = \frac{\pi}{8} C_D \rho_f d_p^2 u_f^2 \quad 2.1$$

Where ρ_f is the fluid density, u_f is the fluid velocity, and C_D is the drag coefficient, which is a function of particles' Reynolds number Re_p . Drag force is predominated by the viscous effect under creeping flow and by the inertia effect in turbulent flow regimes. The drag coefficient under these conditions can be expressed by [2]:

$$C_D = \frac{24}{Re_p} \quad (Re_p < 1) \quad 2.2$$

$$C_D \approx 0.44 \quad (700 < Re_p < 100000) \quad 2.3$$

Considering $Re_p = \rho_f u_f d_p / \mu$ (μ is the fluid viscosity), replacing Eqs. 2.2 and 2.3 in Eq. 2.1 results in:

$$F_D = 3 \pi \mu d_p u_f \quad (Re_p < 1) \quad 2.4$$

$$F_D \approx 0.055 \pi \rho_f d_p^2 u_f^2 \quad (700 < Re_p < 100000) \quad 2.5$$

Eq. 2.4 shows that the drag force is proportional to the fluid viscosity and independent of its density under the viscous flow regime. In contrast, Eq. 2.5 shows that it is proportional to the fluid density and independent of its viscosity under the inertia flow regime.

2.1.2 Buoyancy

The buoyant force is because of the hydrostatic pressure gradient due to the gravitational field. It can be expressed as [3, 4]:

$$F_B = \rho_f V_p g \quad 2.6$$

where V_p is the particle volume, and g is the gravitational acceleration.

Buoyancy is important in liquid-solid fluidization and may become significant in high-pressure gas-solid fluidization. Buoyancy is usually integrated with gravitational force in the form of apparent weight (or buoyant weight).

2.1.3 Gas pressure drop due to HDFs

The pressure of the gas decreases due to the HDFs it exerts on the particles. The Ergun [6] equation is commonly employed to estimate this pressure drop in packed beds. In its general form, the Ergun equation is expressed as follows:

$$\Delta P = C_{Erg} \frac{\mu_g}{d_p^2} \frac{L(1-\varepsilon)^2}{\varepsilon^3} u_f + 1.75 \frac{\rho_g}{d_p} \frac{L(1-\varepsilon)}{\varepsilon^3} u_f^2 \quad 2.7$$

where ε is the bed voidage, L is the bed height and C_{Erg} is a constant with an experimental value of about 150. C_{Erg} decreases when the bed voidage exceeds 0.8 and approaches 72, as its theoretical value for the bed voidage close to one [7].

The first and second terms on the right-hand side of the Ergun equation are referred to as the viscous and inertia terms, respectively, indicating their relative importance in different flow regimes. The ratio between these two terms can be expressed as:

$$\frac{\text{Inertia term}}{\text{Viscous term}} = \frac{Re_p}{85.7(1-\varepsilon)} \quad 2.8$$

Based on Eq. 2.8, the inertia term can be safely neglected at small Reynold's number. This is typically the case in the fluidization of cohesive particles, especially at low gas velocities close to the minimum fluidization conditions. The current study focuses on these conditions, where the viscous effect dominates HDFs. Hence, the inertia term in the Ergun equation is neglected throughout this study.

2.2 Gravitational force

In the absence of IPFs, fluidization is essentially the conditions where the drag force acting on a particle overcomes its apparent weight, W_p , i.e., the resultant force of weight and buoyancy. For a spherical particle W_p can be expressed as follows:

$$W_p = \frac{\pi}{6} d_p^3 (\rho_p - \rho_f) g \quad 2.9$$

where ρ_p is the particle density.

In the fully fluidized state, the gas pressure drop remains relatively constant and equal to the equivalent pressure drop, ΔP_{eq} , i.e., the weight of the particles divided by the bed cross-sectional area:

$$\Delta P_{eq} = \frac{N_p W_p}{A} \quad 2.10$$

where N_p is the total number of particles in the bed, and A is the bed cross-sectional area.

The gas pressure drop is the result of the HDFs acting on particles. Hence, F_D , as the main type of HDFs, is approximately equal to W_p when the bed is fluidized. Therefore, the ratio of *IPFs/HDFs*, which is approximately equal to $IPFs/W_p$, determines the fluidization behaviour of the bed.

2.3 Interparticle forces

Various types of cohesive forces originating from different sources may exist between particles. van der Waals, electrostatic, liquid bridge, and sintering forces are IPFs commonly experienced in fluidization. They will be discussed in the following.

2.3.1 van der Waals forces

The attractive forces resulting from the interactions of the instantaneous and permanent atomic dipoles are called van der Waals forces. The van der Waals forces between two macroscopic particles is the resultant pairwise forces between atomic dipoles in the particles. Many models have been proposed to estimate the van der Waals forces, the simplest of which is the Hamaker [8] model. For the van der Waals forces between two spheres, the Hamaker model is expressed as:

$$F_{vdW} = \frac{H R}{12 a_s^2} \quad 2.11$$

where H is the Hamaker constant as a characteristic of the solid material, R is the radius of the spheres, and a_s is the separation distance. The van der Waals forces are maximally attractive when the separation distance is in the order of molecular diameter, i.e., about 0.4 nm [1].

It became clear that one must consider the local curvature of the particle surface, i.e., its surface asperity size, instead of its radius for a realistic estimation of van der Waals forces by Eq. 2.11 [1, 2]. This is considered in the Rumpf [9] model. According to this model, total van der Waals forces would be the sum of these forces originating from asperity-asperity, asperity-particle, and particle-particle interactions. For two spheres with diameter R and hemispherical asperities of the size r , this model is expressed as:

$$F_{vdW} = \frac{H}{6} \left[\frac{R}{2(a_s + 2r)^2} + \frac{2rR}{(r + R)(a_s + r)} + \frac{r}{2a_s^2} \right] \quad 2.12$$

The ratio of F_{vdW}/W_p decreases with particle diameter as W_p varies with d_p^3 while van der Waals forces are dominated by that surface asperity rather than the particle diameter [1, 10]. This conclusion is the reason behind the more cohesive behaviour of finer particles.

The existence of van der Waals forces in all conditions implies that their magnitude at ambient conditions was implicitly considered in the Geldart classification [11] as it is based on the data of fluidization with air at ambient conditions. The variation of van der Waals forces at elevated temperatures is reflected in the Hamaker constant, which increases virtually linearly with temperature [12]. The growth of these forces is due to an enhanced molecular dipole formation at elevated temperatures. Gas adsorption on the surface of the particles can also contribute to van der Waals forces [13-15] and cause these forces to change with operating pressure and gas type. The magnitude of different types of IPFs depends on the nature of the gas and particle and fluidization conditions. The typical values of IPFs will be discussed in Section 2.4.

2.3.2 Electrostatic force

The electrostatic force is generated when surfaces or particles become electrically charged. In a gas-solid fluidized bed, this happens when non-conducting particles come into contact with each other, the reactor wall, or following electron exchange with the flowing gas. According to Coulomb's law, the electrostatic force between two electrically charged particles is proportional to the product of their electric charge and inversely proportional to the square of the center-to-center distance:

$$F_e = \frac{1}{4 \pi \epsilon} \frac{q_1 q_2}{a^2} \quad 2.13$$

where q_1 and q_2 are the electric charges of the particles, a is the center-to-center distance, and ϵ is the electrical permittivity of the medium.

Compared to van der Waals forces, the electrostatic force is generally insignificant (see Fig. 2.2). In addition, it vanishes in a humid environment and at high temperatures as the particles are discharged by the humidity or as a consequence of an increased conductivity at elevated temperatures [1, 2]. Nonetheless, electrostatic force can result in issues like agglomeration and, in extreme cases, leads to unit shutdown in the fluidization of non-conductive particles, for example, in gas phase polymerization [16].

2.3.3 Liquid bridge

A liquid bridge between adjacent particles can form when there is a small quantity of liquid in the system or if gas moisture content condenses on the surfaces of particles [17]. The bridge generates an attractive force on particles under static and dynamic conditions [1]. The scientific significance of the liquid bridge force is because it can be changed by varying the amount of liquid in the system and liquid properties, i.e., its viscosity and surface tension [18-20]. The liquid bridge force is also important from a practical point of view, for example, in the drying process [1] and high-temperature processing of minerals where impurities combine to create eutectics, i.e., compounds with a low melting point, resulting in the formation of small quantities of liquid in the bed [17].

The dynamic element of liquid bridge force, also known as the viscous force, is generated at the time of a relative movement of the liquid and particle surface [1, 2]. In fluidization, this element

can be neglected in practice, considering that the minimum fluidization velocity of particles is typically smaller than the required relative velocity for the viscous force to be considerable (≈ 1 cm/s for a system with water as the liquid and at a separation distance of about $1 \mu\text{m}$ [1]). According to the boundary method, the static liquid bridge force acting on two spherical particles with radius R can be expressed as [1, 2]:

$$F_l = 2\pi\gamma R \sin \alpha \sin(\alpha + \beta) + \pi R^2 \Delta P \sin^2 \alpha \quad 2.14$$

where γ is the surface tension of the liquid, α and β are characteristic angles shown in Figure 2.1, and ΔP is the pressure difference between the pressure within the liquid and the surrounding environment.

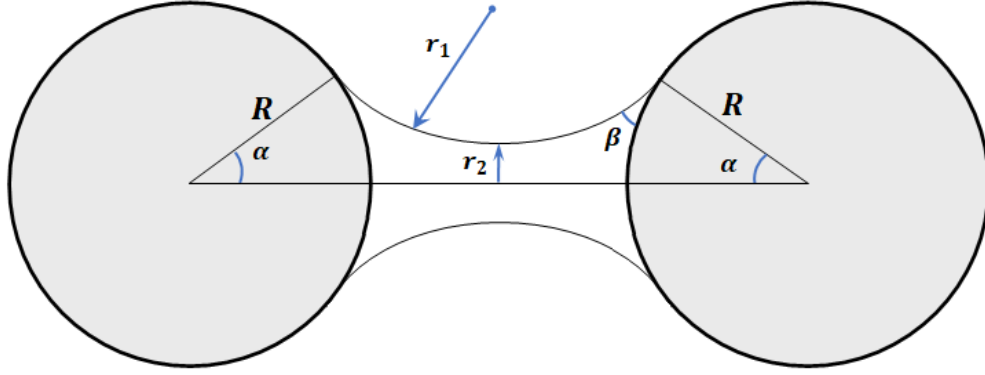


Figure 2.1. Liquid bridge between two spherical particles.

The first and the second terms on the right-hand side of Eq. 2.14 correspond to the liquid surface tension force and pressure deficiency in the liquid, respectively [1, 2]. According to the Laplace-Young equation, ΔP can be expressed as [2]:

$$\Delta P = \gamma \left[\frac{1}{r_1} - \frac{1}{r_2} \right] \quad 2.15$$

The liquid bridge force is highly dependent on the geometry of the bridge and, hence, difficult to be precisely calculated by Eqs. 2.14 and 2.15. Nonetheless, simplifying assumptions can be applied for a straightforward and reasonably precise outcome. According to the gorge method, one can estimate the maximum liquid bridge force by employing the following equation [2]:

$$F_l = 2\pi\gamma R \quad 2.16$$

Like the conditions for estimating van der Waals forces, it is essential to consider the surface asperities in Eq. 2.16 instead of particle radius to obtain a reliable estimate of the liquid bridge force [1, 2].

2.3.4 Sintering

Sintering is the process of forming a solid neck between particles at high temperatures. The cohesion force due to the formation of a neck with radius x can be estimated by [21]:

$$F_s = \pi\sigma_t x^2 \quad 2.17$$

where σ_t is the tensile strength of the neck. The interparticle force that emerges due to sintering is time-dependent and, hence, distinct from other types of IPFs explained earlier. The neck radius grows with time according to an equation with the following form [1]:

$$\left(\frac{x}{R}\right)^2 = Ct \quad 2.18$$

where t is time and C is a constant depending on the material properties and the mechanism of sintering, which can be based on diffusion, viscous flow, and reaction [1, 17]. For viscous sintering, Frenkel proposed the following equation for C [22]:

$$C = \frac{3\gamma}{2R\mu} \quad 2.19$$

Eq. 2.19 indicates that viscous sintering is driven by the surface energy and resisted by the viscosity. γ is a weak function of temperature, whereas μ decreases sharply with temperature. Consequently, increasing the temperature of a gas-solid fluidized bed above the minimum sintering temperature of particles can enhance the sintering process and significantly increase the level of IPFs [17]. The sintering process is enhanced at higher temperatures regardless of the sintering mechanism. Severe sintering leads to the formation of larger agglomerates and might eventually

lead to bed defluidization [23]. Sintering is commonly experienced in gas phase polymerization, combustion, gasification, reduction of iron, and high-temperature processing of low-quality minerals containing impurities with low sintering temperatures [1, 17].

2.4 Magnitudes of different types of IPFs

Typical magnitudes of pairwise van der Waals, electrostatic, liquid bridge, and sintering forces are compared with the weight of a particle in Figure 2.2. The calculation details and approximated parameters are presented in Table 2.1.

Table 2.1. Details of calculation and parameters for the estimation of different forces in Figure 2.2.

Force	Equation	Parameters and remarks
Weight of a particle	$W_p = \frac{\pi}{6} d_p^3 \rho_p g$	➤ $1000 \text{ kg/m}^3 < \rho_p < 3000 \text{ kg/m}^3$
van der Waals	Eq. 2.11: $F_{vdw} = \frac{H R}{12 a_s^2}$	➤ R was replaced by asperity size ($r = 0.1 \mu\text{m}$). ➤ $1 \times 10^{-20} \text{ J} < H < 1 \times 10^{-19} \text{ J}$. ➤ $a_s = 4 \times 10^{-10} \text{ m}$. ➤ Maximum value of F_{vdw} is estimated by Eq. 2.11 as a was set to the average molecular diameter.
Electrostatic	Eq. 2.13: $F_e = \frac{1}{4 \pi \epsilon} \frac{q_1 q_2}{a^2}$	➤ $\epsilon = 8.85 \times 10^{-12} \text{ F/m}$. ➤ $q_1 = q_2 = \delta A_p$ where A_p is the particle surface area ($A_p = \pi d_p^2$) and δ is the charge surface density. ➤ $2 \times 10^{-7} \text{ C/m}^2 < \delta < 1.4 \times 10^{-6} \text{ C/m}^2$. ➤ $a \approx d_p$ (=center to center distance).
Liquid bridge	Eq. 2.16: $F_l = 2\pi\gamma R$	➤ R was replaced by asperity size ($r = 0.1 \mu\text{m}$). ➤ $4 \times 10^{-2} \text{ N/m} < \gamma < 4 \times 10^{-1} \text{ N/m}$. ➤ Maximum value of F_l is estimated by Eq. 2.16.

Sintering	Eq. 2.17: $F_s = \pi \sigma_t x^2$	➤ F_s was estimated as if x equals the asperity size ($r = 0.1 \mu m$). ➤ $1 \times 10^7 Pa < \sigma_t < 1 \times 10^8 Pa$.
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Figure 2.2 demonstrates that, for particles smaller than about $1000 \mu m$, sintering and liquid bridge forces are greater than other types of IPFs. While the maximum magnitude of the liquid bridge force was calculated and presented in Figure 2.2, it is important to recognize that sintering can further increase as the neck grows. The magnitude of the sintering force and the possibility of its growth explain why sintering can lead to defluidization even in beds of large particles.

Liquid and solid bridge forces may exist only under special circumstances. For instance, these forces are not present in a gas-solid system operating at ambient conditions in a dry environment. Under such circumstances, van der Waals forces, which are always present in a gas-solid system, take over. Figure 2.2 shows that van der Waals forces become greater than W_p for particles smaller than $30\text{-}100 \mu m$. This is supported by the observations indicating that IPFs acting on particles smaller than this size range commonly resist gravity. The presented range of van der Waals forces in Figure 2.2 corresponds to the typical values of the Hamaker constant at ambient conditions. The Hamaker constant is greater at higher temperatures [12]. Consequently, van der Waals forces become more significant under these circumstances.

Figure 2.2 shows that, unlike material bridge and van der Waals forces, the electrostatic force increases with particle diameter. The rate of the increase of this force is, however, lower than the increase in W_p . Electrostatic force becomes comparable with W_p for particles smaller than about $30 \mu m$. Nevertheless, van der Waals forces are greater than electrostatic forces for these powders. Hence, electrostatic force is typically negligible in comparison with van der Waals forces or W_p . However, the electrostatic force can become considerable for non-conductive particles, e.g., polymers [16], owing to their capacity for higher electrical charge density.

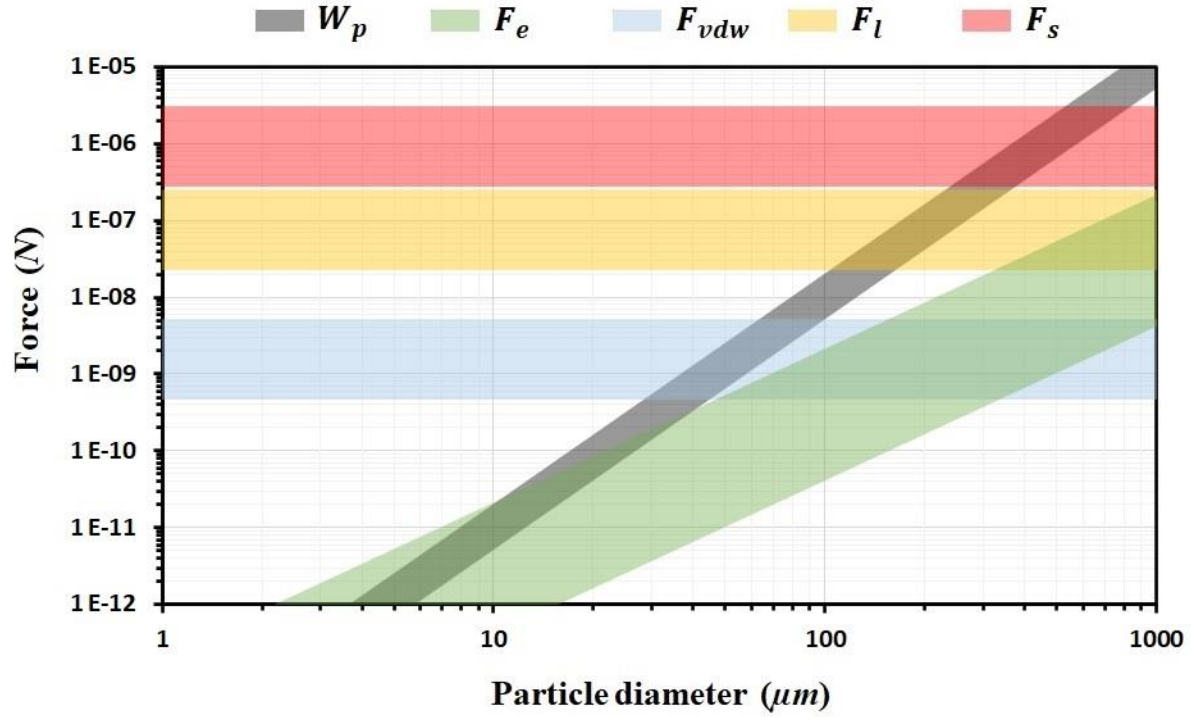


Figure 2.2. Typical magnitudes of pairwise electrostatic (F_e), van der Waals (F_{vdw}), liquid bridge (F_l), and sintering (F_s) forces, in comparison with the weight of a particle (W_p). Refer to Table 2.1 for calculation details.

2.5 Effect of interparticle forces on the hydrodynamics of gas-solid fluidized beds

After introducing the Geldart classification, it became evident that the fluidization behaviour of Geldart group A and, more notably, group C are significantly influenced by IPFs [24, 25]. At this step, let us limit our discussion to fluidization at ambient conditions in a dry environment, as Geldart proposed his classification under these circumstances [11], where liquid bridge and sintering forces do not exist [2]. The major contribution to the total magnitude of IPFs is, hence, van der Waals forces. Under these conditions, d_p of particles with Geldart group C behaviour is typically smaller than about $30\ \mu\text{m}$. This particle diameter coincides with the lower bound of the range where pairwise van der Waals forces become greater than W_p (see Figure 2.2). Therefore,

considering the coordination number of particles, total IPFs acting on a particle of Geldart group C must be at least about an order of magnitude greater than W_p . Hence, IPFs acting on these powders can defy gravity and govern the fluidization behaviour of these powders. As a result, the fluidization characteristics of these powders, e.g., channeling and plugging [11], are closely linked to their cohesivity. On the other end of the spectrum, there exist large and heavy particles with Geldart groups B and D behaviours. The size of these particles, typically larger than $100 \mu m$, implies that W_p is considerably greater than IPFs acting on these particles (see Figure 2.2). Between the above two extremes, there exist Geldart group A particles for which IPFs and W_p must be approximately in the same order of magnitude. Geldart classification is presented in Figure 2.3, where schematics of relative magnitudes of forces acting on particles in different groups are also shown.

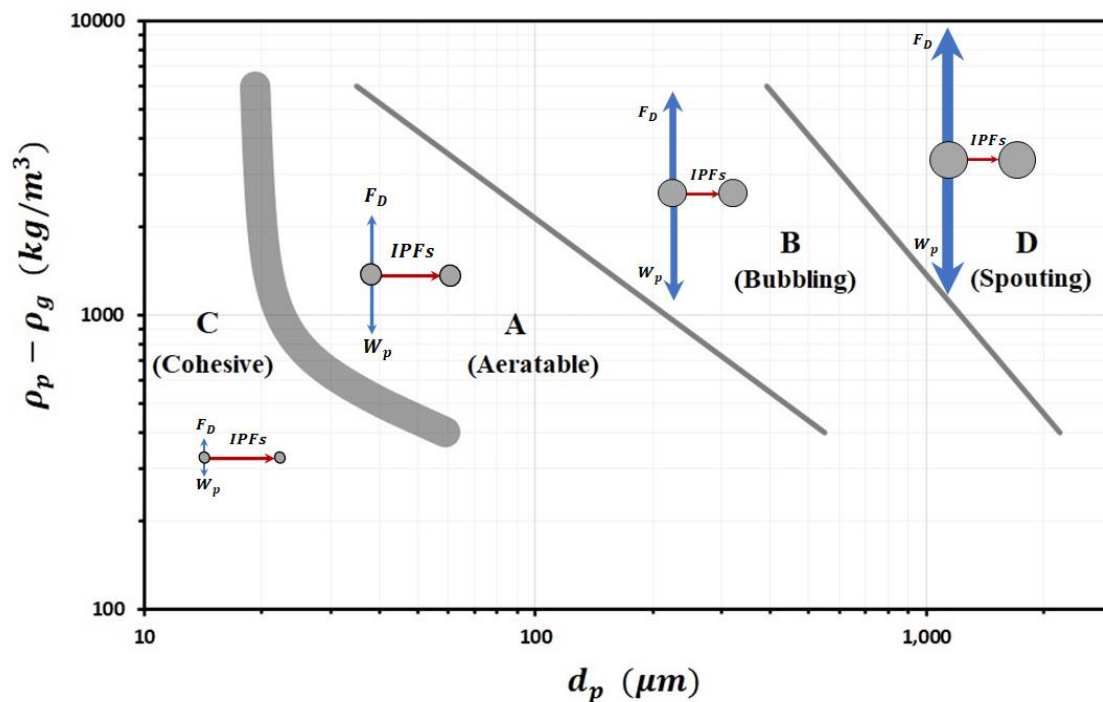


Figure 2.3. Geldart classification and relative magnitude of forces acting on particles.

Geldart's diagram assigns a group label to a given particulate assembly based on the particle diameter and the density difference, $\rho_p - \rho_g$. As can be predicted from the above remarks, however, the fluidization behaviour of a given particulate system can shift by changing the ratio of $IPFs/W_p$ upon variation of IPFs [17]. Accordingly, a particulate assembly must show different Geldart group behaviours based on the variation of IPFs. This conclusion broadly agrees with experimental results in literature [20, 26-33], where various techniques have been employed to change the level of IPFs. They include increasing operating temperature [26-28], application of a magnetic field on beds of magnetic particles [29, 30], the addition of a non-volatile liquid [32], experiments with various moisture content [20], surface treatment techniques like polymer coating [31], the addition of a small amount of nano-sized particle that stick to the surface of host particles [33], and employing water as the fluidizing medium [20]. In all cases, Geldart group behaviour of particles shifted from group D and/or B towards group A and/or C following a growth in the level of IPFs [20, 26-32] and vice versa [20, 33].

As alluded to earlier, a significant change in the level of IPFs may transform the Geldart group behaviour of a particulate system. However, a mild alteration of IPFs can change the bed's hydrodynamic parameters while the general Geldart group behaviour remains unchanged [27, 31, 34, 35]. In the following, the effects of IPFs on the most important hydrodynamic parameters for the design and scale-up of a gas-solid fluidized bed are discussed.

IPFs increase the gas flow required to fluidize the particles [17, 31, 36]. This leads to an increase in minimum fluidization velocity [27, 28, 31, 37, 38], minimum bubbling velocity [25, 27, 39, 40], and transition velocity from bubbling to turbulent fluidization regime [31, 35]. Hence, less gas remains available for bubbling activity leading to a decrease in bubble velocity and passage frequency [31, 35], granular temperature [41], and solid entrainment rate [31], and an increase in the particle's idle time [42]. The bed void fraction also increases with IPFs. This includes bed voidage at loose, minimum fluidization, and minimum bubbling conditions, as well as emulsion phase voidage [27, 34, 35, 43]. These modifications have two opposing effects on the bed hydrodynamics and, subsequently, on the bed performance. On the one hand, the increase in the interstitial gas velocity and emulsion phase voidage enhances the gas-solid contact and, subsequently, the bed performance [44]. On the other hand, the quality of solid mixing decreases [42, 45] and agglomerates form following a substantial increase in the level of IPFs [41, 46]. These

modifications enhance the gas screening effect [47], i.e., the tendency of the gas to pass through intra-agglomerate spaces rather than inter-agglomerate void. The bed performance reduces upon diminished gas-solid contact because of the latter effects. Therefore, a mild increase in the level of IPFs seems to boost the beneficial effects of IPFs while avoiding the detrimental ones, including bed defluidization in extreme cases.

2.6 Measurement of IPFs

Several experimental approaches have been proposed for measuring IPFs or qualitatively evaluating their variation. These techniques can be categorized into three groups: (i) measurement of IPFs between individual particles, (ii) measurement of IPFs based on rheological properties of the bulk, and (iii) measurement of IPFs in a gas-solid fluidized bed. These approaches are reviewed in this section, and their accuracy and applicability to gas-solid fluidized beds are discussed.

2.6.1 Measurement of IPFs between individual particles

Several techniques are commonly used to measure interparticle forces between two individual particles. They include atomic force microscopy (AFM), optical tweezers, application of centrifugal force, and inverse application of discrete element method (DEM).

Atomic force microscopy: AFM is commonly employed to investigate surface topology [48, 49]. It uses a sharp probe, typically a cantilever with a tiny tip, that scans the surface of a sample. To measure IPFs with AFM, a particle is affixed to the probe tip. As the tip approaches the other particles, IPFs cause a deflection in the cantilever. By monitoring this deflection, the interparticle forces can be quantified. AFM provides precise measurements of IPFs at different separation distances [50-52].

Optical tweezers: In this technique, microscopic particles are trapped and manipulated by the momentum of the photons of highly focused laser beams [53]. IPFs can be calculated by analyzing the position and movement of two trapped particles close to each other. Optical tweezers are widely employed to investigate biological systems, DNA, and protein molecules due to their capability to

manipulate single molecules and real-time response in dynamic processes [54]. Like AFM, the optical trapping technique can accurately measure IPFs between microscopic particles at different separation distances [53].

Application of centrifugal force: Centrifuges were used in early studies to estimate adhesion forces between particles and a surface. The principle is, however, applicable to two particles. At first, centrifugal force is applied to press a particle against the surface of interest or another particle attached to the surface. The surface is then reversed, and the centrifugal force required to detach the particle is calculated based on the rotational speed at which the particle is separated [49].

Liquid bridge force measurement: Gray et al. [55] presented a novel device for measuring pull-off force between particles generated by liquid bridging of vacuum residue at high temperatures. In this apparatus, particles are affixed to the end of two rods. One of these rods is fixed, while the second can move and is connected to a linear variable differential pressure transducer. The force required to separate the particles is then correlated with the voltage produced by the differential pressure transducer.

Inverse application of discrete element method: DEM is a numerical technique commonly used to simulate the behaviour of particulate systems. This method is typically used to iteratively predict the position of particles with time, given the forces acting on them. DEM can also be inversely applied to quantify IPFs from experimental data of the particles' position and movement. Hence, employing this technique requires precise knowledge of the particles' trajectory [56].

The above techniques have been significantly improved in recent years, specifically AFM and Optical tweezers which are currently among the most accurate methods for measuring IPFs between individual particles. However, the particles in a bulk solid have diverse characteristics, like shape, size, and surface properties, that affect IPFs. Consequently, conducting many tests becomes necessary to obtain a representative IPF for a particulate system. In addition, IPFs measured by AFM and Optical tweezers are a function of the separation distance between particles [50-53]. In a particulate system, this separation distance is uncertain. It varies from particle to particle and under different conditions, for instance, at different gas velocities in a gas-solid fluidized bed. Thus, researchers commonly investigate the bulk properties to indirectly evaluate IPFs in particulate systems.

2.6.2 Measurement of IPFs with the rheological properties of the bulk

Due to the limitations in applying the techniques described in Section 2.6.1 to a bulk solid, it is preferable to measure the rheological properties of a particulate system rather than IPFs between individual particles, particularly when specific conditions like temperature or moisture must be controlled [49]. These techniques are discussed in this section.

Shear test and torque measurement: The shear test involves subjecting a particulate sample to controlled normal and shear forces while monitoring the corresponding flow response [49]. In a typical shear test, a sample is placed between two parallel plates or within a shear cell under certain levels of normal stress. The applied shear stress is then gradually increased until the sample begins to flow. As shown in Figure 2.4, the yield shear stress against normal stress usually follows a straight line called yield limit [49]. When extrapolated, the yield limit of non-cohesive particles passes the origin, which suggests that these particles flow freely. The tensile strength is defined as the normal stress at zero shear stress and estimated by extrapolating the yield limit. For cohesive particles, extra shear stress is required to overcome the tensile strength of the bulk with a non-zero value because of IPFs [52, 57-59].

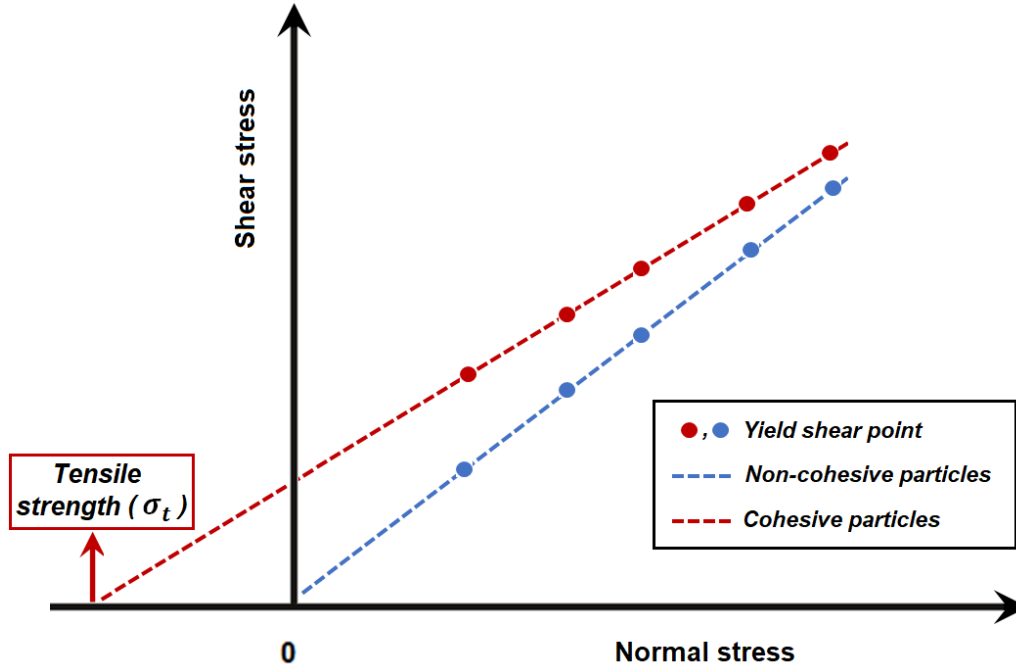


Figure 2.4. Typical results of a shear test for cohesive and non-cohesive particles.

Molerus [60] proposed the following relationship between the normal force on a particle surface, F_N , and the normal stress applied on the sample bulk solid, σ :

$$F_N = \frac{\pi d_p^2}{(1 - \varepsilon)} \sigma \quad 2.20$$

Under zero shear stress, i.e., at $\sigma = \sigma_t$, this equation is assumed to quantify the IPFs acting on a particle [52, 58, 59]:

$$IPFs = \frac{\pi d_p^2}{(1 - \varepsilon)} \sigma_t \quad 2.21$$

Measurement of torque required to rotate a surface submerged in a solid bulk is another technique to investigate the flow properties of a particulate system. This technique is similar to the shear test in principle [49, 61]. Torque measurement technique can be applied to different conditions of the particulate assembly, including its fluidized state [61].

The shear test is one of the most common and mature experiments to determine a particulate bulk's tensile strength and flow behaviour. This technique is extensively used when solid handling is important, for example, in the pharmaceutical and soil-related fields [49]. The shear test cannot, however, replicate the conditions of the particulate assembly in a gas-solid fluidized bed. This is due to the nature of this experiment, where solid bulk is sheared under different levels of normal stress that decrease the specimen voidage to values smaller than that in the loose state. Deducing IPFs from a shear test also involves extrapolating the results to the points of zero shear stress, which was shown to be associated with a high degree of uncertainty, especially for more cohesive powders [49]. Moreover, the Molerus equation, which is commonly used for the estimation of IPFs with the results of a shear test, was originally developed to calculate the force transmitted by different layers of particles [60]. Hence, this equation's applicability for quantifying IPFs by replacing normal stress with tensile strength is questionable.

Characteristic angles: Particulate materials make different angles depending on their nature and physical properties. They include the angle of repose, which can be measured for poured and drained particles, and the angle of slide or gravity chute angle, which can be measured in static and dynamic modes. The latter is sometimes called the dynamic angle of repose or avalanching angle. Characteristic angles are shown in Figure 2.5. Among other parameters like particle size and shape, characteristic angles are influenced by the balance between gravitational force, IPFs, and friction between particles [49, 50, 59, 62-64].

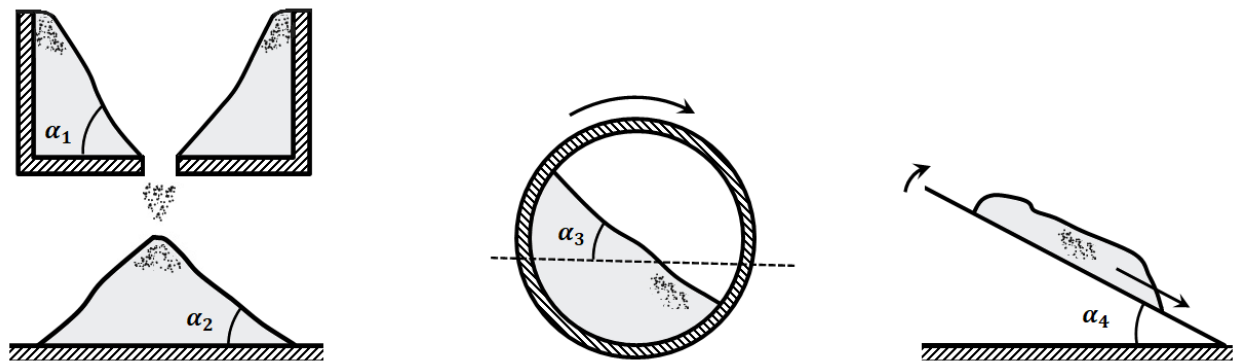


Figure 2.5. Characteristic angles: α_1 : angle of repose (drained), α_2 : angle of repose (poured), α_3 : angle of slide (dynamic), and α_4 : angle of slide (static).

Baerns [63] proposed a quantitative approach to estimate IPFs and the friction between particles by the angle of slide. The method is based on measuring the static angle of slide with a surface covered with a layer of particles glued to it. At the angle of slide, the force of gravity overcomes the friction and IPFs, i.e.:

$$mg \sin \alpha_4 = \mu_f mg \cos \alpha_4 + IPFs \times N \quad 2.22$$

where m is the mass of the specimen, μ_f is the friction coefficient, and N is the number of particles in the sliding area, approximated by dividing the area to d_p^2 . By measuring the angle of slide for different specimen masses and plotting $mg \sin \alpha_4$ against $mg \cos \alpha_4$, a linear relationship is observed. $IPFs$ and μ_f are subsequently estimated by the intercept and the slope of this line, respectively.

Characteristic angles are primarily important in the design of solid handling apparatus like silos and expansion zone of fluidized beds. Measuring these angles for cohesive powders involves a significant level of uncertainty due to their tendency to form irregular shaped surfaces [49]. Hence, information on the characteristic angles is often evaluated in combination with other methods for a comprehensive understanding of the particles' flow behaviour rather than quantifying IPFs [50, 64, 65].

The application of tilting fluidized beds while observing the deviation of the bed surface from a horizontal position is analogous to the analysis of the characteristic angles. This technique can be employed to qualitatively evaluate the level of IPFs in a gas-solid fluidized bed [66].

Hausner ratio: Hausner ratio (HR) is defined as the ratio of the tapped and loose bulk densities, which can also be considered as the ratio of the solid phase fractions of the tapped and loose states:

$$HR = \frac{\rho_{tap}}{\rho_0} = \frac{1 - \varepsilon_{tap}}{1 - \varepsilon_0} \quad 2.23$$

For non-cohesive particles, loose and tapped solid phase fractions are close since these particles can easily flow without applying an external force. Hence, HR is close to one for non-cohesive particles. Cohesive particles, on the other hand, tend to have higher interlocking. Consequently, a greater force is required for these particles to flow and rearrange. As a result, the solid phase

fraction increases more significantly when this force is applied by tapping. This implies that the HR is greater for cohesive particles. It was suggested that HR is about 1.4 at the transition from Geldart groups A to C [25].

HR is an index for a qualitative evaluation of IPFs, while no quantitative relationship between the two is currently available. HR can be conveniently measured under various operating conditions. However, the test procedure, like the time and type of tapping (manual/automatic), can affect its measurement [49, 64].

Other qualitative tests: Many experiments have been proposed to evaluate the flowability and rheology of particulate materials. The results of these experiments for cohesive powders are also considered qualitative indications of the magnitude of IPFs. These experiments include the following:

- Flow function: Defined as the ratio of consolidation stress applied to a specimen to the corresponding unconfined yield strength. A smaller flow function is associated with greater cohesivity [49].
- Funnel test: Measuring discharge time in funnels with different outlet diameters.
- Sieving test: Measuring the remaining mass of powders on a specific sieve as a function of time.
- Penetration test: Measuring the required force for inserting a pointed probe into the solid bulk.
- Carr compressibility index: Similar to the Hausner ratio with the following definition [49]:

$$K_I = \frac{\rho_{tap} - \rho_0}{\rho_0} \times 100 \quad 2.24$$

2.6.3 Measurement of IPFs in a gas-solid fluidized bed

Despite the crucial importance of IPFs in fluidization, only a few approaches have been proposed to quantify these forces under the conditions of a particulate assembly in a gas-solid fluidized. These approaches are discussed in this section.

Fluidizability index: Baerns [63] measured IPFs for several cohesive powders by their angles of slide and compared the results with the fluidizability index (FI) which he defined as the ratio of theoretical prediction for the minimum fluidization velocity ($U_{mf,pred}$) to its experimental value ($U_{mf,exp}$):

$$FI = \frac{U_{mf,pred}}{U_{mf,exp}} \quad 2.25$$

Experimental results showed that a greater level of IPFs is associated with a smaller fluidizability index. This remark agrees with findings in the literature showing that $U_{mf,exp}$ increases with IPFs [27, 28, 31, 37, 38]. Based on the value of FI , Baerns [63] identified the following fluidization behaviour zones: normal fluidization ($FI > \sim 10^{-1}$), quasi-fluidization ($\sim 10^{-3} < FI < \sim 10^{-1}$), where channels form at $U_{mf,exp}$ but transform into bubbles by increasing the gas velocity, and no fluidization zone ($FI < \sim 10^{-3}$).

The qualitative correlation between FI and IPFs indicates the possibility of developing in-situ approaches to quantify IPFs in a gas-solid fluidized bed. This is one of the achievements of the current study, i.e., the U_{mf} deviation equation, which is presented in Chapters 4 and 5.

Measurement of electrostatic charge: In a gas-solid fluidized bed, electrostatic charge is commonly quantified by measuring particle charge-to-mass ratio, electric current, or the electric potential induced by charged particles [16]. A Faraday cup can be employed to directly measure the charge density of a particulate specimen. In principle, this device consists of two connected metal cups, an inner and an outer one, forming a Faraday cage where electrical charges move to the outer surface. Particles are poured into the inner cup, and an electrometer measures the current between the cups [16]. Mehrani et al. [67] developed a novel fluidized bed Faraday cup for in-situ measurement of the charge density in a gas-solid fluidized bed. Accumulated charge in fluidized beds can also be indirectly measured by sensors and probes, including capacitance, induction, and

collision probes [16]. Like the inverse DEM approach for measuring the force between two particles, the electrostatic charge of a single particle can be calculated by precise information on its trajectory in an electromagnetic field [16].

The above techniques, however, may not be appropriate for estimating total IPFs if other types of IPFs are important. This includes common scenarios such as the fluidization of fine powders and high-temperature operation.

Bed pressure drop in a vibrated expanded bed: Jaraiz et al. [68] developed an experimental approach coupled with a theoretical analysis to estimate IPFs for cohesive powders that cannot be fluidized without an assisting method. This approach is based on the variation of the bed pressure drop in the homogeneous fluidization state generated by the bed vibration. The model assumes that the increase in the bed pressure drop with gas velocity in the homogeneous regime is due to the breakage of agglomerates with the gas flow. This increases bed voidage and decreases coordination number. Based on this model, pairwise IPFs can be estimated by the following equation [68]:

$$\text{Pairwise IPFs} = \frac{\pi d_p^3}{6L_0(1 - \varepsilon_0)} \times \left(-\frac{d(\Delta P_b)}{d(k)} \right) \quad 2.26$$

where L_0 is the bed height at ε_0 , ΔP_b is the bed pressure drop, and k is the coordination number which is usually considered a mere function of the bed voidage. Eq. 2.26 correlates IPFs to the slope of the line representing pressure drop vs coordination number.

The model does not consider the effects of vibration intensity and frequency on the breakage of agglomerates [68] and, hence, on the variation of pressure drop and coordination number. In addition, this approach applies only to cohesive particles that cannot be fluidized without an assisting method.

Sound-assisted fluidization: An acoustic field can break the agglomerates and assist in fluidizing cohesive powders. Russo et al. [69] proposed the cluster/subcluster oscillator model for describing the sound-assisted fluidization of cohesive powders. In this model, the size of agglomerates is determined by the balance of IPFs, and breakage forces, i.e., forces originating from the sound wave and gas flow. Chirone et al. [70] and Raganati et al. [37] applied this model to estimate IPFs for cohesive powders. The results showed that the estimated IPFs highly depend on the sound

frequency and intensity [37, 70]. This is caused by the cluster/subcluster oscillator model not considering sound attenuation [69]. Like the previously explained approach employing bed vibration, the sound-assisted fluidization approach only applies to cohesive powders with Geldart group C behaviour.

Overshoot in the bed pressure drop profile at U_{mf} : The appearance of an overshoot at U_{mf} in the bed pressure drop profile when the gas velocity is gradually increased is considered an indication of the presence of IPFs [31, 71, 73]. Some researchers suggested that the pressure drop overshoot required for the fluidization of cohesive particles is equal to the tensile strength of the particulate bed [71-73]. Accordingly, IPFs are estimated by the adapted Molerus equation [60], Eq. 2.21. However, the pressure drop overshoot might be partially caused by the friction between particles and the bed wall [73, 74]. In addition, the bed pressure drop and, subsequently, the pressure drop overshoot is proportional to the bed height [6, 71], while the tensile strength and IPFs are intrinsic properties of particles. Hence, applying Eq. 2.21 results in calculating IPFs as a function of the bed height [71], which can be viewed as a fundamental problem of this approach.

Hysteresis model: Recently, Affleck et al. [75] proposed the hysteresis model, which is also based on the pressure drop overshoot at U_{mf} and can be considered as a modification to the non-generalized DHR (dynamic Hausner ratio) approach which was developed in the current study (see Chapter 4). The non-generalized DHR equation quantifies IPFs based on the difference between ε_{mf} and ε_0 as the origin of the appearance of the pressure drop overshoot and addressed the issues of the proposed approaches based on the overshoot pressure drop, i.e., their dependence on the bed height. The non-generalized DHR equation, however, has limitations in quantifying IPFs under agglomerating fluidization. The Hysteresis model is an attempt to overcome this limitation by incorporating the variation in the coordination number Δk during the bed expansion from ε_0 to ε_{mf} into the DHR approach. The results of this model can be expressed as [75]:

$$\frac{IPFs}{W_p} = \frac{\Delta P_{overshoot}}{f \Delta P_{eq} \Delta k} \quad 2.27$$

where f is the fluidization quality index defined as the ratio of the bed pressure drop at high gas velocities and its equivalent pressure drop, ΔP_{eq} .

Compared with the non-generalized DHR approach, the hysteresis model quantified more realistic values of IPFs for fine particles that form agglomerates. However, it led to virtually similar values of IPF/W_p for particles in the range of about 30 – 150 μm , while it is expected to be significantly smaller for larger particles.

2.7 Incorporation of IPFs in Geldart classification

As discussed previously, the magnitude of IPFs is in the same order of that of W_p in the case of Geldart group A behaviour, while IPFs play a dominant role in the fluidization behaviour of group C powders. Consequently, the criteria proposed for transitioning between these two groups are either directly based on the magnitude of IPFs or on parameters influenced by these forces, such as the Hausner ratio and angle of repose (AOR). In contrast, IPFs are relatively insignificant compared to HDFs acting on particles with Geldart groups B and D behaviours. Hence, most correlations and criteria proposed for distinguishing between these groups are based on parameters or dimensionless numbers that represent hydrodynamic forces, mainly by Archimedes number (Ar), which is defined as the ratio of gravitational and viscous forces, and dimensionless density, i.e., $(\rho_p - \rho_g)/\rho_g$ [76-79]. The proposed criteria based on the magnitude of IPFs for the transition between different Geldart groups are listed in Table 2.2, where the corresponding equivalent IPF/W_p are also presented.

Table 2.2. Proposed criteria based on the magnitude of IPFs for the boundary between Geldart groups A and C (A-C) and between Geldart groups A and B (A-B).

	Reference	Criteria	Equivalent IPF/W_p (If applicable)
A-C	Baerns [63] a	$\frac{W_p}{W_p + IPFs} = 5 \times 10^{-2}$	$\frac{IPFs}{W_p} = 19$

	Molerus [24] b	$10 \frac{(\rho_p - \rho_g) d_p^3 g}{F_{vdw}} = 10^{-2}$	$\frac{IPFs}{W_p} \approx 5800$
	Chaouki et al. [80]	$U_g > U_t$ at $F_D = W_p + IPFs$ (or $U_{mf,exp} > U_t$)	—
	Geldart et al. [25]	$HR = 1.25$ to 1.4	—
	Zhao et al. [81]	$AOR = 35^\circ$ to 40°	—
	Wang [82]	$\hat{S}_e = \frac{1}{d_p(\rho_p - \rho_g)} > 25$	—
	Mostoufi [79] c	$\frac{IPFs}{W_p} = \frac{50}{Ar^{0.583}} + 0.71$	$\frac{IPFs}{W_p} \approx 100$
A-B	Molerus [24] d	$\frac{(\rho_p - \rho_g) \left(\frac{\pi}{6} d_p^3\right) g}{F_{vdw}} = 0.16$	$\frac{IPFs}{W_p} \approx 33$
	Mostoufi [79] e	$\frac{IPFs}{W_p} = \frac{20}{Ar^{0.662}} + 0.07$	$IPFs/W_p \approx 5.3$

- a) The criterion corresponds to the quasi-fluidization conditions [63] (see Section 2.6.3).
- b) The equivalent $IPFs/W_p$ was estimated considering that the resultant IPFs acting on a particle is equal to the product of F_{vdw} and coordination number (Eq. 6.15). The coordination number was estimated by Eq. 6.16 assuming $\varepsilon_{mf} = 0.65$.
- c) The equivalent magnitude of IPFs is the average value for typical powders close to the Geldart A-C boundary ($1000 < \rho_p < 3000 \text{ kg/m}^3$, $15 < d_p < 30 \mu\text{m}$, fluidization with air at ambient conditions).
- d) The same procedure as b assuming $\varepsilon_{mf} = 0.45$.
- e) The same procedure as c assuming $1000 < \rho_p < 3000 \text{ kg/m}^3$, $70 < d_p < 210 \mu\text{m}$.

To estimate the magnitude of IPFs, studies listed in Table 2.2 adopted different approaches. For instance, Baerns [63] evaluated the total IPFs acting on a particle by the angle of slide, Molerus [24] estimated van der Waals forces by the Krupp model [83], and Mostoufi [79] calculated total

IPFs by theoretical estimations of different types of IPFs. Consequently, the proposed magnitudes of IPFs corresponding to the A-C boundary vary by orders of magnitude ($IPFs/W_p = 19$ [63], 5800 [24], and 100 [79]). A similar difference between the proposed $IPFs/W_p$ for Geldart A-B boundary is observed in Table 2.2 ($IPFs/W_p = 33$ [24], and 5.3 [79]). In addition, proposed $IPFs/W_p$ for the A-B boundary are substantially greater than one, whereas IPFs are expected to be insignificant compared to W_p for Geldart group B behaviour. The extensive range of the proposed $IPFs/W_p$ for the Geldart A-C and A-B boundaries and their unrealistic values imply that the adopted approaches for the quantification of IPFs are not applicable to the conditions of a gas-solid fluidized bed.

2.8 Nomenclature

Acronyms

A-B	boundary between Geldart groups A and B
A-C	boundary between Geldart groups A and C
AFM	atomic force microscopy
AOR	angle of repose
DEM	discrete element method
DHR	dynamic Hausner ratio
FI	fluidizability index
HDFs	hydrodynamic forces
HR	Hausner ratio
IPFs	interparticle forces

Symbols

A	bed cross-sectional area (m^2)
Ar	Archimedes number (-)
a	center-to-center distance between two particles (m)
a_s	separation distance between two particles (m)
C	constant in Eq. 2.18 ($1/\text{s}$)
C_D	drag coefficient (-)
C_{Erg}	constant in the Ergun equation, Eq. 2.7 (-)
d_b	bubble diameter (m/s)
d_p	particle diameter (m)
F_B	force of buoyancy (N)
F_D	drag force (N)
F_l	liquid bridge force (N)
F_N	normal force on the surface of a particle (N)
F_s	sintering force (N)
F_{vdw}	van der Waals forces (N)
f	fluidization quality index (-)
g	gravitational acceleration (m^2/s)
H	Hamaker constant (J)
$IPFs$	interparticle forces acting on each particle (N)
k	coordination number (-)
L	bed height (m)
L_0	bed height in loose conditions (m)

m	mass of the sliding specimen (Kg)
N	number of particles in the sliding area (-)
N_p	total number of particles in the bed (-)
q_1	electrical charge of particle #1 (C)
q_2	electrical charge of particle #2 (C)
R	particle radius (m)
Re_p	particle Reynolds number (-)
r	asperity size (m)
r_1	radius of the curvature of liquid bridge in Figure 2.1 (m)
r_2	radius of the neck of the liquid bridge in Figure 2.1 (m)
$\hat{S}_e$	specific surface area index (m ² /Kg)
t	time (s)
U_b	bubble rise velocity (m/s)
u_f	fluid velocity (m/s)
U_{mf}	minimum fluidization velocity (m/s)
$U_{mf,exp}$	experimental minimum fluidization velocity (m/s)
$U_{mf,pred}$	minimum fluidization velocity predicted by the Carman-Kozeny equation (m/s)
V_p	volume of a particle (m ³)
W_p	weight of a single particle (N)
x	radius of the sintering neck (m)

Greek letters

α	angle in Figure 2.1 (-)
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α_1	drained angle of repose (-)
α_2	poured angle of repose (-)
α_3	dynamic angle of slide (-)
α_4	static angle of slide (-)
β	angle in Figure 2.1 (-)
γ	surface tension (N/m)
Δk	difference in coordination number at ε_0 and ε_{mf} (-)
ΔP	pressure difference between within the liquid and the surrounding environment (Pa)
ΔP_b	bed pressure drop (Pa)
ΔP_{eq}	equivalent pressure drop (Pa)
ΔP_{mf}	bed pressure drop at minimum fluidization conditions (Pa)
$\Delta P_{overshoot}$	pressure drop overshoot at minimum fluidization (Pa)
ϵ	electrical permittivity ($C^2/(Nm^2)$)
ε	voidage (-)
ε_{mf}	minimum fluidization voidage (-)
ε_{tap}	tapped bulk voidage (-)
ε_0	loose bulk voidage (-)
μ	viscosity (Pa. s)
μ_f	friction coefficient (-)
μ_g	gas viscosity (Pa. s)
ρ_f	fluid density (kg/m^3)

ρ_g	gas density (kg/m ³)
ρ_p	particle density (kg/m ³)
ρ_{tap}	tapped bulk density (kg/m ³)
ρ_0	loose bulk density (kg/m ³)
σ	normal stress applied on the sample bulk solid (Pa)
σ_t	tensile strength (Pa)

CHAPTER 3 COHERENCE OF THE CHAPTERS

Chapter 2 of the thesis provided a literature review on interparticle forces and their impact on the hydrodynamics of gas-solid fluidized beds, where the measurement techniques for evaluating the magnitude of these forces and their applicability to the conditions of a gas-solid fluidized bed were comprehensively discussed. It was shown that IPFs can significantly affect the hydrodynamics of a gas-solid fluidized bed, especially at higher temperatures, where the magnitude of IPFs is typically greater than that at ambient conditions and for fine and ultrafine powders with typical behaviour of Geldart groups A and C. These situations are commonly experienced in industrial gas-solid fluidized beds. For instance, most catalytic gas-solid fluidized bed reactors use fine powders showing Geldart group A behaviour. In addition, the use of ultrafine particles and nanopowders is growing due to their large surface area and numerous applications. Moreover, industrial fluidized beds are commonly operated at elevated operating conditions. The bed hydrodynamics and its Geldart behaviour cannot adequately be predicted by conventional models, which are based purely on hydrodynamic principles and disregard IPFs.

The impact of IPFs on the bed hydrodynamics and, subsequently, on its design and scale-up procedure, operation, and overall performance, highlights the significance of an accurate estimation of the magnitude of these forces. As demonstrated in Chapter 2, the available techniques for measuring IPFs generate unrealistic and inconsistent results as they are hardly applicable to the conditions of particulate materials in gas-solid fluidized beds. Hence, it is necessary to develop reliable in-situ approaches to effectively quantify IPFs under these circumstances. These approaches can subsequently be employed to include IPFs in the Geldart classification of particles and correlations for estimating hydrodynamic parameters.

Considering the points mentioned above, the main objective of this research study is to "develop a fundamental basis for the quantitative investigation of IPFs in a gas-solid fluidized bed and their effects on the bed hydrodynamics".

The following specific objectives were defined to achieve the main objective:

- Develop in-situ approaches to quantify IPFs and their variation with operating conditions in a gas-solid fluidized bed.
- Extension of Geldart classification to consider the effect of IPFs.

The accomplishment of these objectives is presented in Chapters 4, 5, 6, and 7, which include three scientific articles and a supplementary chapter.

Chapter 4 is dedicated to developing two approaches for in-situ quantification of the magnitude of IPFs and its variation with temperature in a gas-solid fluidized bed. For the determination of the absolute values of IPFs, the "DHR (Dynamic Hausner ratio)" equation was developed. The " U_{mf} (minimum fluidization velocity) deviation" approach was devised to estimate thermally generated IPFs, i.e., the difference between IPFs at a given temperature and ambient conditions.

The limitations of the proposed approaches in Chapter 4 include an underestimation of the magnitude of IPFs by the DHR approach in gas agglomerating fluidization and incapability of the U_{mf} deviation approach in quantifying the absolute magnitude of IPFs. These limitations were addressed in Chapter 5 by introducing generalized U_{mf} deviation and DHR equations.

The generalized equations for in-situ quantification of IPFs proposed in Chapter 5 were employed in Chapter 6 to extend Geldart classification by considering the effect of IPFs.

Findings in Chapter 6 include the variation of the magnitude of IPFs with bed voidage. A model was presented in Chapter 7 to establish quantitative relationships between the bed voidage, agglomerate size, and magnitude of IPFs.

The thesis concludes by presenting a general discussion in Chapter 8, highlighting the key advancements achieved through this research, and providing recommendations for future works in Chapter 9.

CHAPTER 4 ARTICLE 1 : IN-SITU QUANTIFICATION OF THE MAGNITUDE OF INTERPARTICLE FORCES AND ITS TEMPERATURE VARIATION IN A GAS-SOLID FLUIDIZED BED

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Published on 7 December 2020, Chemical Engineering

Science DOI: <https://doi.org/10.1016/j.ces.2020.116349>

Highlights:

- Quantification of the magnitude of interparticle forces and its temperature variation in a gas-solid fluidized bed.
- Quantification of thermally induced interparticle forces based on the difference between the experimental and theoretical minimum fluidization velocities.
- Increase in the dynamic Hausner ratio, i.e., the ratio of the minimum fluidization voidage to the loose bulk voidage, with interparticle forces.
- An equation for the quantification of interparticle forces by the dynamic Hausner ratio.

4.1 Abstract

Interparticle forces (IPFs) are among the important parameters that can influence the hydrodynamics of a gas-solid fluidized bed and, hence, its overall performance. They are particularly critical at elevated temperatures, where their magnitude is typically greater than that at ambient conditions. Two simple and indirect approaches were developed in this study to quantify the magnitude of IPFs and its variation with temperature in a gas-solid fluidized bed. Quantification

of thermally induced IPFs, i.e., the difference between the level of IPFs at a given temperature and that at ambient conditions, was attempted in the first approach. It was shown that the variation of IPFs with temperature is related to the difference between the experimental minimum fluidization velocity ($U_{mf,exp}$) and that predicted by a correlation ($U_{mf,pred}$) based purely on hydrodynamic principles. The magnitude of thermally induced IPFs at a given temperature was then quantified with the help of the Ergun equation and the difference between $U_{mf,exp}$ and $U_{mf,pred}$. The second approach was developed for the quantification of absolute values of IPFs based on the difference between the loose bulk voidage (ε_0) and the minimum fluidization voidage (ε_{mf}). It was shown that the magnitude of IPFs directly affects the ratio of $\varepsilon_{mf}/\varepsilon_0$, which we called “dynamic Hausner ratio”. A phenomenological model was, hence, proposed where the magnitude of IPFs is expressed as a function of the dynamic Hausner ratio. The performance of these approaches were verified and qualitatively validated, while their limitations and opportunities were also discussed.

Keywords: Gas-solid fluidized bed, Hydrodynamics, Interparticle forces, Quantification, High temperature, Dynamic Hausner ratio.

4.2 Introduction

Gas-solid fluidized beds are broadly employed in chemical and metallurgical industries due to their unique features, including the favorable heat and mass transfer characteristics, excellent gas-solid contact, uniform and controllable temperature, and ability to handle a wide variety of particulate materials [1-4]. Interparticle forces (IPFs) may considerably affect the hydrodynamics of a gas-solid fluidized bed [5-9] and, consequently, its overall performance [5, 10]. Representative examples, where IPFs principally govern the bed hydrodynamics, include the fluidization of fine powders with typical Geldart [11] group C behaviour [12-14] and agglomerating fluidization owing to the presence of an enhanced level of cohesive liquid and/or solid bridge IPFs in the bed [15-17]. Therefore, the successful design, scale up, operation, and performance evaluation of gas-solid fluidized beds are greatly dependent on the comprehensive understanding of IPFs and their impact on the bed hydrodynamics [3, 5, 8].

The magnitude of IPFs and its relative importance in comparison with gravitational and hydrodynamic forces (HDFs), i.e., the forces that originate from fluid-particle interaction, determine their influence on the bed hydrodynamics [10, 12, 18]. Some researchers recommended that the boundaries between different Geldart group behaviours correspond to certain ratios of IPFs to the particle weight [12, 19]. This is in broad agreement with the findings reported in the literature showing that upon increasing the level of cohesive IPFs, a fluidized bed of particles with typical Geldart group B behaviour, which is dominated by HDFs [12], can show characteristics of Geldart group A or even group C [5, 10, 20, 21]. Such substantial modifications in the bed hydrodynamics highlight the critical effect of IPFs and underline the necessity to consider them for a thorough prediction of the bed hydrodynamics.

The level of IPFs can be considerably affected by the operating temperature [10, 22]. The variation of IPFs with temperature is critically important since most industrial fluidized beds operate at elevated temperatures [6, 23, 24]. In addition, the application of gas-solid fluidized beds at high temperature is expected to become more routine in the near future owing to the rarefaction of conventional resources and increased tendency to process lower quality feedstocks, like waste materials [10]; a condition where the bed may be operated under an enhanced level of IPFs.

The main types of IPFs in a particulate system are van der Waals, electrostatic, and material bridge forces, which can form due to either the liquid or solid bridge [10, 13, 25]. The level of electrostatic forces decreases with temperature [26, 27] owing to an enhanced electrical conductivity of particles at higher temperatures [28]. However, despite some exceptions, e.g., in a wet bed where water induced liquid bridge forces vanish upon evaporation of water at temperatures slightly higher than ambient conditions, the level of other cohesive IPFs generally increases with temperature [20, 22, 29]. Although the level of electrostatic forces decreases with temperature [26, 27] owing to an enhanced electrical conductivity of particles at higher temperatures [28], the level of other cohesive IPFs generally increases with temperature [20, 22, 29]. The growth of IPFs is particularly significant when approaching the sintering temperature of particles [10, 30] or upon the development of liquid bridge between particles due to the formation of eutectics at high temperature [10, 13, 25]. Disregarding the effect of temperature on the level of IPFs is likely one of the principal reasons behind the considerable controversy in the literature on the effect of temperature on bed hydrodynamics [10, 20]. This, in turn, recommends that the exclusive

application of hydrodynamic correlations, which merely account for the variation of gas properties with temperature, cannot adequately predict the bed behaviour at elevated temperatures [10, 31]. The main challenge in adequately accounting for the contribution of IPFs in hydrodynamic correlations lies in the unavailability of a reliable technique for the quantification of IPFs in a fluidized bed, in particular, at elevated temperatures.

Experimental approaches for the evaluation of IPFs can be categorized into three groups: force measurement between individual particles, approaches based on the measurement of powders' rheological characteristics, and fluidization approaches, as presented in Table 4.1. Each of these approaches has its own advantages and limitations that make it suitable for specific applications. The first and second categories are advantageous owing to their high level of accuracy and simplicity, respectively. However, the structure of an aerated particulate assembly in a gas-solid fluidized bed, which is normally composed of particles with different properties, each in a specific configuration/distance with its surrounding particles, cannot be replicated by these approaches. The same challenge is encountered when models and equations proposed for the theoretical prediction of IPFs are applied in a fluidized bed. Only a limited number of approaches have been proposed for in-situ measurement of IPFs in a gas-solid fluidized bed categorized under the fluidization approaches in Table 4.1. These approaches are, in general, incapable of adequately quantifying IPFs, particularly for the powders with typical Geldart group A behaviour that are commonly employed for catalytic reactions. In this study, two simple and indirect approaches are presented to quantify the magnitude of IPFs and its variation with operating temperature in a gas-solid fluidized bed. The performances of these approaches were verified and qualitatively validated.

Table 4.1. Experimental approaches for the qualitative/quantitative evaluation of interparticle forces.

Category	Approach	Concept	Remarks/ limitations
Force measurement approaches between individual particles	Centrifugal force approach [32]	Adopting centrifugal force to separate particles	• AFM and optical trapping are among the most precise approaches. • Applicable for two or a limited number of particles and not for a particulate assembly, e.g., one in a gas-solid fluidized bed. • Accurate measurement of particle-particle distance or particles' trajectory is required. • Liquid bridge force measurement was only applied on two rods covered with a thin liquid film. It is likely to be applicable for coarse particles, e.g., with a few millimetres in diameter, and not for fine particles.
	Optical trapping approach [33]	Manipulating particles with a light beam	
	Atomic force microscopy (AFM) [34-36]	Deflection of the microscope cantilever	
	Liquid bridge force measurement [37]	Measurement of pull-off force with a pressure transducer	
	Discrete element method (DEM) [38]	Inverse application of DEM method by knowing the particles' trajectories	
Rheological approaches	Shear test/torque measurement [22, 32, 36, 39-41]	Required shear stress for incipient flow of particles	• IPFs may be quantified by some of these approaches, e.g., shear test [22, 42] or angle of slide [18, 43]. However, most of them are qualitative. • Results are highly dependant on the test procedure [32, 44]. • Bulk structure conditions in these tests are different from its condition in a fluidized bed [45, 46]. Thus, they are not appropriately applicable to a gas-solid fluidized bed.
	Hausner ratio [34, 41, 44, 47]	The ratio of tapped to loose bulk densities	
	Characteristic angles [18, 34, 41, 43, 44]	Different angles that a particle assembly make	
	Other flowability tests, e.g., penetration test, Carr flowability index, etc. [32, 41]		

Fluidization approaches	Fluidizability index [43]	The ratio of the minimum fluidization velocity (U_{mf}) calculated by a conventional correlation ($U_{mf,pred}$) to its experimental value ($U_{mf,exp}$)	• Qualitative
	Electrostatic force measurement [48]	Measurement of electrostatic charge by faraday cups or electrostatic probes/sensors	• Applicable for quantification of electrostatic forces only and not other IPFs, which are usually significant at high temperature.
	Bed pressure drop in a vibrated expanded bed [49]	Variation of the bed pressure drop with particles' coordination number	• Solely applicable for fine cohesive powders with typical Geldart group C behaviour. • Effects of vibration intensity and frequency on the breakage of agglomerates are not considered in the proposed model.
	Sound assisted fluidization [45]	Application of an acoustic field for the breakage of agglomerates	• Solely applicable for fine cohesive powders showing typical Geldart group C behaviour. • The results depend on the sound frequency and intensity [45].
	Overshoot in the bed pressure drop profile at U_{mf} [46, 50, 51]	Assuming that the tensile strength of particles is identical to the bed pressure drop overshoot at U_{mf} , and consequently applying the equation proposed by Rumpf [52] and Molerus [42] to quantify IPFs	• The pressure overshoot might be partially due to the wall friction [46, 53]. • The bed pressure drop is proportional to the bed height [50, 54], while the tensile strength and IPFs are intrinsic properties of particles. Hence, application of Rumpf [52] and Molerus [42] equation results in the calculation of IPFs as a function of the bed height [50].

4.3 Method development

4.3.1 U_{mf} deviation approach

Minimum fluidization velocity is among the most critical fluidization characteristics of powders. It is defined as the point where the total bed pressure drop reaches a plateau corresponding to the equivalent bed pressure drop, i.e., the weight of particulate bed divided by the bed cross-sectional area [1]. The effect of temperature on the minimum fluidization velocity has been the subject of many studies [10, 30]. Operating temperature affects U_{mf} by one or both of the following drifts: (i) alteration of the gas properties, which influences HDFs [10, 30], and (ii) changing the level of IPFs [5, 6, 20, 29]. Purely hydrodynamic based correlations, such as Carman–Kozeny [55] and Ergun [54] equations, are commonly adopted for the prediction of minimum fluidization velocity. Although these correlations account for the effect of gas properties, they cannot adequately predict U_{mf} if the level of IPFs is appreciable [6, 20, 29]. For instance, Formisani et al. [29] and Botterill et al. [56] showed that Carman–Kozeny and Ergun equations underestimate U_{mf} for relatively fine particles at elevated temperatures, when merely adjusting the gas properties. They attributed this observation to the variation of minimum fluidization voidage with temperature as a result of the thermal growth in the level of IPFs [29, 56]. Similarly, Baerns [43] showed that the fluidizability index (refer to Table 4.1) is closely related to IPFs. The delayed U_{mf} under an enhanced level of IPFs can be ascribed to the need for a greater flow of gas to overcome IPFs in addition to the particles' weight [5, 57].

The deviation of $U_{mf,exp}$ from $U_{mf,pred}$ at a given temperature forms the foundation of the first approach developed in this study for the quantification of thermally induced IPFs, i.e., the difference between the level of IPFs at a given temperature and that at ambient conditions. $U_{mf,pred}$ in this approach must be estimated by a correlation that is capable of accurately predicting $U_{mf,exp}$ at ambient conditions, while adequately considering the effect of gas properties. We will show in Section 4.6.1 how the Carman-Kozeny equation can satisfy these criteria. The

additional bed pressure drop originating from the thermally induced IPFs at a given temperature, $\Delta P_{\Delta IPFs}$, can be quantified by the Ergun equation [54] as follows:

$$\begin{aligned} \Delta P_{\Delta IPFs} = 150 \frac{\mu_g}{(\phi_s d_p)^2} \frac{L_{mf}(1 - \varepsilon_{mf})^2}{\varepsilon_{mf}^3} (U_{mf,exp} - U_{mf,pred}) \\ + 1.75 \frac{\rho_g}{\phi_s d_p} \frac{L_{mf}(1 - \varepsilon_{mf})}{\varepsilon_{mf}^3} (U_{mf,exp}^2 - U_{mf,pred}^2) \end{aligned} \quad 4.1$$

where μ_g and ρ_g are the gas viscosity and density, L_{mf} , and ε_{mf} are the bed height and voidage at minimum fluidization conditions, and d_p and ϕ_s are particle diameter and shape factor, respectively. L_{mf} and ε_{mf} must be considered at their corresponding values at ambient conditions as the effect of temperature will be merely accounted for on the gas properties in Eq. 4.1.

The second term on the right-hand side of Eq. 4.1 becomes important for coarse particles [30] and can be safely neglected when the viscous effect dominates over inertia, i.e., for fine and intermediate-sized particles for which the particle Reynolds number at U_{mf} ($Re_{p,mf} = \frac{\rho_g d_p U_{mf}}{\mu_g}$) is less than 20 [1]. The difference between total IPFs at a given temperature and that at ambient conditions, $\Delta IPFs_{total}$, is equal to the additional force that is required to overcome excess IPFs at that temperature with the gas flow. It can be estimated as:

$$\Delta IPFs_{total} = 150 \frac{\mu_g}{(\phi_s d_p)^2} \frac{A L_{mf}(1 - \varepsilon_{mf})^2}{\varepsilon_{mf}^3} (U_{mf,exp} - U_{mf,pred}) \quad , \quad Re_{p,mf} < 20 \quad 4.2$$

where A is the bed cross-sectional area. Assuming that the bed is made of identical powders, each having a particle weight of W_p , the ratio of thermally induced IPFs acting on a single particle to the particle weight can, thus, be calculated by:

$$\frac{\Delta IPFs}{W_p} = \frac{\Delta IPFs_{total}}{N_p W_p} \quad 4.3$$

where N_p is the total number of particles in the bed. The product of $N_p W_p$ is the total weight of the bed. Hence:

$$\frac{\Delta IPFs}{W_p} = 150 \frac{\mu_g}{(\phi_s d_p)^2} \frac{AL_{mf}(1 - \varepsilon_{mf})^2}{m_s g \varepsilon_{mf}^3} (U_{mf,exp} - U_{mf,pred}) \quad , \quad Re_{p,mf} < 20 \quad 4.4$$

where m_s is the bed inventory and g is the gravitational acceleration. By substituting $\frac{m_s}{AL_{mf}(1 - \varepsilon_{mf})}$ with particle density, ρ_p , Eq. 4.4 can be reformulated as:

$$\frac{\Delta IPFs}{W_p} = \frac{150}{(\phi_s d_p)^2} \frac{1 - \varepsilon_{mf}}{\varepsilon_{mf}^3} \frac{\mu_g}{\rho_p g} (U_{mf,exp} - U_{mf,pred}) \quad , \quad Re_{p,mf} < 20 \quad 4.5$$

where ρ_p is the particle density. As alluded to earlier, while the corresponding values of μ_g , $U_{mf,exp}$, and $U_{mf,pred}$ at the desired temperature are employed in Eq. 4.5, ε_{mf} must be considered at its value at ambient conditions. Eq. 4.5 can be adopted to quantify the ratio of the thermally induced IPFs acting on each particle to its weight at a given temperature. This approach, which we refer to as the “ U_{mf} deviation approach”, is, however, incapable of quantifying the absolute values of IPFs. This was the principal motivation for the development of a second approach in this study, which will be described in the next section.

4.3.2 Dynamic Hausner ratio approach

4.3.2.1 Background and assumptions

The gas flowing through a packed bed of particles with negligible IPFs must principally overcome the weight of the particles for their fluidization. Therefore, the particles can start their movements, determined as their minimum fluidization point, with minimal modification in their packing structure as soon as the force exerted by the flowing gas exceeds their weight. This implies that ε_{mf} must be similar to the fixed bed voidage, ε_0 , for these particles. Hence, the total bed pressure drop profile reaches a plateau after a linear increase with the superficial gas velocity without

showing an overshoot at U_{mf} . This is a typical behaviour of Geldart group B and/or D particles. However, when the bed is made of powders with typical Geldart group A and/or C behaviour, the flowing gas needs to exert a force in excess of the particles' weight to overcome IPFs and fluidize the particles. Therefore, at superficial gas velocities lower than U_{mf} in the increasing velocity pass, where the bed is at ε_0 , the particles are kept stagnant at their places by both IPFs and their weight. When the gas velocity reaches U_{mf} , HDFs become equal to the sum of IPFs and the weight of particles. The bed voidage then experiences a sudden increase from ε_0 to ε_{mf} and IPFs are weakened by increasing the average mutual particle-particle distance (see Supplementary Material). Consequently, particles are allowed to move and be fluidized. The sudden increase in the bed voidage at minimum fluidization conditions coincides with the sharp decrease in the bed pressure drop in the increasing velocity pass. In the decreasing pass, however, the bed voidage gradually decreases from ε_{mf} to ε_0 with the decrease in the gas velocity from U_{mf} to zero, leading to a smooth transition from the fluidized to fixed bed state without any pressure overshoot. The above discussion recommends that (i) the difference between the bed pressure drop profiles in the increasing and decreasing velocity passes at U_{mf} corresponding to the difference between ε_0 and ε_{mf} originates from IPFs, and (ii) the level of IPFs acting on particles at ε_{mf} is considerably lower than that at ε_0 .

The pressure overshoot might be partially attributed to the wall friction [46, 53]. However, it will be shown in Section 4.6.2 that the difference between ε_0 and ε_{mf} and, hence, the corresponding pressure overshoot are certainly caused by IPFs. This forms the basis of the second approach for quantifying IPFs in a gas-solid fluidized bed. The following general assumptions were adopted for the development of a model for the quantification of IPFs based on this approach:

- (i) The particulate bed is made of powders identical in physical characteristics, e.g., size, sphericity, and material.
- (ii) The minimum fluidization velocity is determined from the bed pressure drop profile in the decreasing velocity pass.

- (iii) The difference between the total bed pressure drops in the increasing and decreasing velocity passes at U_{mf} , i.e., $(\Delta P_{U_{mf}, \varepsilon_0} - \Delta P_{U_{mf}, \varepsilon_{mf}})$ in Figure 4.1 is equal to the required additional pressure drop to decrease the level of IPFs from what is experienced at ε_0 to that at ε_{mf} .
- (iv) The total bed pressure drops at U_{mf} when the bed is at ε_0 and ε_{mf} can be calculated with the viscous term of the Ergun equation. Therefore, the model being developed here will be applicable for cases with $Re_{p,mf} < 20$, where the viscous effect dominates.
- (v) A small fraction of IPFs acts on the particles even when the bed is at ε_{mf} . To take this into account in the model, we assume that some particles are not fluidized under this condition, e.g., those particles in the near-wall area or dead zones. Thus, IPFs acting on these particles are equal to IPFs at the fixed bed state. The remaining particles are assumed to be individually fluidized, while no IPFs act on them. With this strategy, the fraction of IPFs that are overcome during the bed expansion from ε_0 to ε_{mf} would be equal to the fraction of fluidized particles at U_{mf} , represented by $F_{fp,mf}$.
- (vi) If the flowing gas exerts any force on the particles, it experiences a pressure drop when passing through the bed. If all particles were fluidized, the total bed pressure drop would have been equal to the equivalent pressure drop, ΔP_{eq} . The ratio of the total bed pressure drop at the fluidized state to ΔP_{eq} was introduced as the fluidization quality index and was shown to decrease by increasing the powder cohesivity [58, 59]. Applying the same concept, $F_{fp,mf}$ in assumption (v) is postulated to be equal to the ratio of $\Delta P_{U_{mf}, \varepsilon_{mf}}$ to ΔP_{eq} as follows:

$$F_{fp,mf} = \frac{\Delta P_{U_{mf}, \varepsilon_{mf}}}{\Delta P_{eq}} = \frac{\Delta P_{U_{mf}, \varepsilon_{mf}}}{m_s g / A} \quad 4.6$$

4.3.2.2 Model development

According to assumption (iii), total IPFs that are overcome when the bed voidage increases from ε_0 to ε_{mf} , $IPFs_{total}$, can be expressed as:

$$IPFs_{total} = \left(\Delta P_{U_{mf}, \varepsilon_0} - \Delta P_{U_{mf}, \varepsilon_{mf}} \right) A \quad 4.7$$

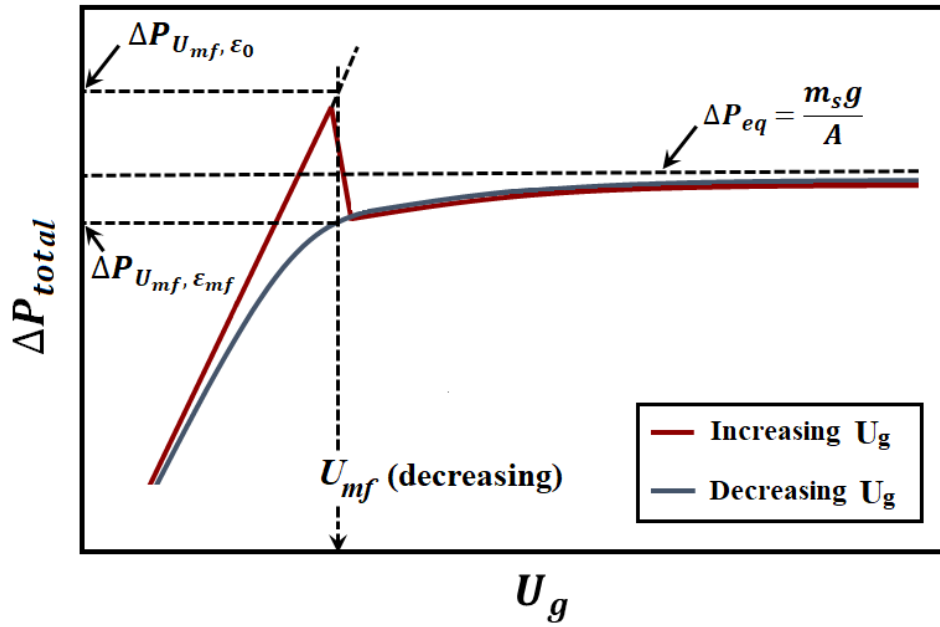


Figure 4.1. Schematic representation of a typical bed pressure drop profile for Geldart group A behaviour.

By dividing $IPFs_{total}$ by the number of fluidized particles, the magnitude of IPFs acting on each particle in their loose bulk state can be quantified as:

$$IPFs = \frac{\left(\Delta P_{U_{mf}, \varepsilon_0} - \Delta P_{U_{mf}, \varepsilon_{mf}} \right) A}{F_{fp, mf} N_p} \quad 4.8$$

By dividing both sides of Eq. 4.8 by the weight of a particle, we have:

$$\frac{IPFs}{W_p} = \frac{(\Delta P_{U_{mf}, \varepsilon_0} - \Delta P_{U_{mf}, \varepsilon_{mf}})A}{F_{fp, mf} N_p W_p} = \frac{(\Delta P_{U_{mf}, \varepsilon_0} - \Delta P_{U_{mf}, \varepsilon_{mf}})A}{F_{fp} m_s g} \quad 4.9$$

According to assumption (iv), $\Delta P_{U_{mf}, \varepsilon_0}$ and $\Delta P_{U_{mf}, \varepsilon_{mf}}$ can be estimated as:

$$\Delta P_{U_{mf}, \varepsilon_0} = \frac{150}{(\phi_s d_p)^2} \frac{L_0 (1 - \varepsilon_0)^2}{\varepsilon_0^3} \mu_g U_{mf} \quad 4.10$$

$$\Delta P_{U_{mf}, \varepsilon_{mf}} = \frac{150}{(\phi_s d_p)^2} \frac{L_{mf} (1 - \varepsilon_{mf})^2}{\varepsilon_{mf}^3} \mu_g U_{mf} \quad 4.11$$

where L_0 and L_{mf} as the bed heights at loose bulk and minimum fluidization states are related owing to the bed inventory conservation as follows:

$$L_{mf} (1 - \varepsilon_{mf}) = L_0 (1 - \varepsilon_0) \quad 4.12$$

By substituting Eq. 4.11 for $\Delta P_{U_{mf}, \varepsilon_{mf}}$ in Eq. 4.6, $F_{fp, mf}$ can be calculated as:

$$F_{fp, mf} = \frac{150}{(\phi_s d_p)^2} \frac{A L_{mf} (1 - \varepsilon_{mf})^2 \mu_g U_{mf}}{\varepsilon_{mf}^3 m_s g} \quad 4.13$$

By substituting Eqs. 4.10-4.13 into Eq. 4.9 and rearranging the resulted equation, we obtain:

$$\frac{IPFs}{W_p} = \left(\frac{\varepsilon_{mf}}{\varepsilon_0} \right)^3 \frac{1 - \varepsilon_0}{1 - \varepsilon_{mf}} - 1 \quad 4.14$$

Eq. 4.14 can be adopted for quantifying IPFs in a gas-solid fluidized bed, while it is only a function of ε_{mf} and ε_0 . The ratio of $IPFs/W_p$ calculated by Eq. 4.14 is plotted against $\varepsilon_{mf}/\varepsilon_0$ for different values of ε_{mf} in Figure 4.2. It shows that the magnitude of IPFs has a greater functionality in $\varepsilon_{mf}/\varepsilon_0$ than only ε_{mf} or ε_0 . For example, over the explored span of $\varepsilon_{mf}/\varepsilon_0$ and ε_{mf} in Figure 4.2, an approximate change of 8% in the $IPFs/W_p$ is obtained at a given $\varepsilon_{mf}/\varepsilon_0$ for a 10% variation in ε_{mf} . However, a 10% change in $\varepsilon_{mf}/\varepsilon_0$ around 1.2 at a given ε_{mf} results in about a 50% change

in the calculated $IPFs/W_p$, while the change is more significant for smaller $\varepsilon_{mf}/\varepsilon_0$. This is in accordance with the experimental results presented in Section 4.6.2.

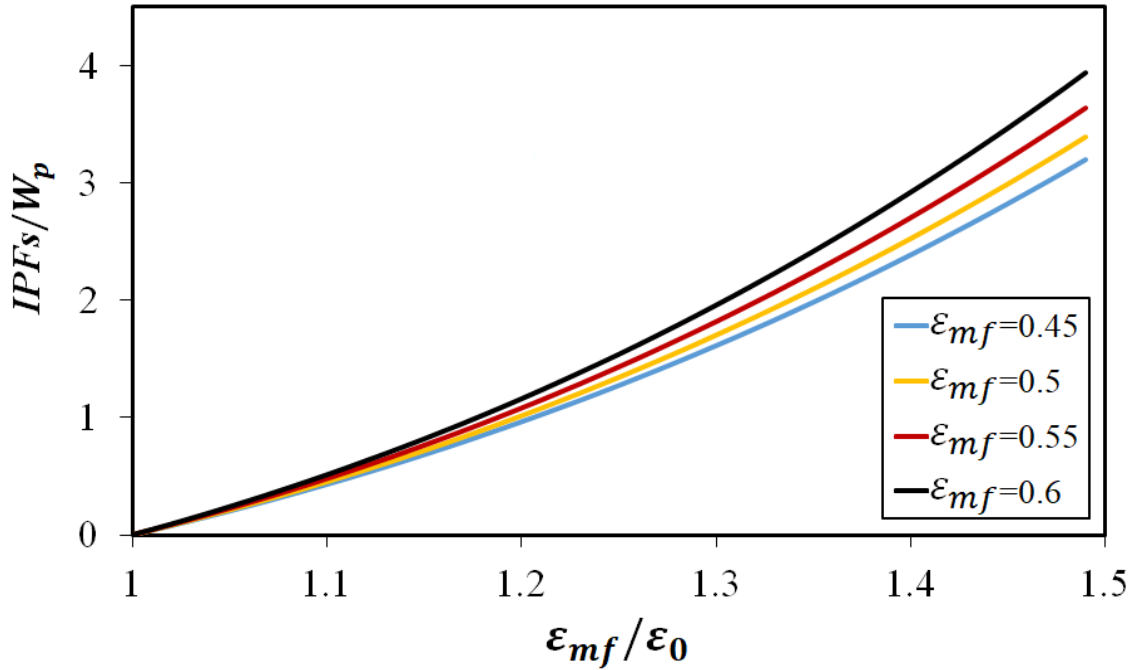


Figure 4.2. $IPFs/W_p$ as a function of $\varepsilon_{mf}/\varepsilon_0$ and ε_{mf} based on Eq. 4.14 (for Geldart group A).

The Hausner ratio (HR), as the ratio of tapped to loose bulk densities, is commonly employed for a qualitative evaluation of IPFs and prediction of fluidization behaviour [5, 34, 41, 58, 60]. The ratio of $\varepsilon_{mf}/\varepsilon_0$ and its relationship with IPFs based on Eq. 4.14 shows similarities to HR as listed below:

- Both HR and $\varepsilon_{mf}/\varepsilon_0$ are always equal to or greater than one.
- The magnitude of IPFs is negligible when $\varepsilon_{mf}/\varepsilon_0 \sim 1$ based on Eq. 4.14. This is usually the case for intermediate-sized and/or coarse particles showing typical Geldart group B and/or D behaviour for which no or a very small pressure overshoot can be identified in the bed

pressure drop profile (see Figure 4.5). Likewise, HR is approximately one for these particles [5, 60].

- According to Eq. 4.14, the magnitude of IPFs increases with $\varepsilon_{mf}/\varepsilon_0$. Similarly, HR is higher for a system with a greater level of IPFs [5, 34, 60].
- Refer to Figure 4.2, $IPFs/W_p$ is approximately equal to one when $\varepsilon_{mf}/\varepsilon_0$ is about 1.2-1.25 and IPFs tend to be dominant at higher values of $\varepsilon_{mf}/\varepsilon_0$. Therefore, if $\varepsilon_{mf}/\varepsilon_0$ is approximately less than 1.2-1.25, the powders are expected to show typical Geldart group A behaviour for which IPFs and particle weight are in the same order of magnitude [12, 49]. This is in accordance with the results reported in the literature for particles showing typical Geldart group A behaviour [61] and those obtained in this study (see Figure 4.6). Likewise, a particulate assembly is expected to demonstrate typical behaviour of Geldart group A if $1.0 < HR < 1.2-1.25$ [34, 58, 60].
- If $HR > 1.4$, particles are expected to behave like Geldart group C [34, 58, 60] for which IPFs govern the fluidization behaviour [12-14]. Figure 4.2 also shows that IPFs quantified by Eq. 4.14 are considerably greater than the particle's weight if $\varepsilon_{mf}/\varepsilon_0 > 1.4$.

Based on the above discussion, we called the ratio of $\varepsilon_{mf}/\varepsilon_0$ “dynamic Hausner ratio” (DHR) owing to its similarity to HR and the dynamic nature of minimum fluidization conditions, where ε_{mf} and, hence, DHR are measured. The name of the current approach for the quantification of IPFs was, thus, inspired by this terminology.

Both U_{mf} deviation and DHR approaches quantify the resultant IPFs acting on each particle by all surrounding particles rather than the mutual particle-particle adhesive forces. This is justifiable and even favorable in a gas-solid fluidized bed, where the behaviour of particles is affected by the resultant IPFs as in-bed particles are always in the environment of other particles [43]. Nevertheless, knowing the resultant IPFs acting on each particle, one can estimate the mutual particle-particle adhesive forces considering the structural configuration of particulate assembly and its corresponding coordination number.

4.4 Verification and validation

The performances of U_{mf} deviation and DHR approaches to accurately quantify the magnitude of IPFs and its variation with temperature in a gas-solid fluidized bed was verified and qualitatively validated in this study. The former was accomplished by monitoring the magnitude of quantified IPFs, as well as the independency of these approaches to the gas type (for the adopted gases and solid materials under the experimental conditions explored in this study) and bed dimensions. The qualitative validation was achieved through the comparison of the temperature variations of the magnitude of quantified IPFs and normalized peak area in the bed pressure drop profile with respect to its value at ambient conditions.

Magnitude of IPFs in a gas-solid fluidized bed: The relative importance of IPFs in comparison with gravitational and/or hydrodynamic forces is different for particles with different Geldart group characteristics. For particles with typical Geldart group B behaviour, IPFs are negligible compared to their weight, while they are in the same order of magnitude for particles behaving like Geldart group A [12, 19]. Quantified IPFs for particles showing typical behaviours of Geldart groups A and B were analyzed with these criteria.

Effect of gas type on quantified IPFs: The fluidizing gas can contribute to the level of van der Waals forces through adsorption on the surface of particles [62-64]. Gas adsorption is highly dependent on the gas and particle physical properties [63, 64] and is enhanced at elevated pressures [62]. The explored operating conditions in the current study include ambient pressure and/or high temperature, where there is negligible gas adsorption on the surface of particles for the gases and solids adopted in this study [63, 64]. This implies that the quantified IPFs in the different gaseous environments under operating conditions explored in this study must be similar.

Quantification of IPFs in beds with different dimensions: IPFs are among the intrinsic characteristics of particles and, hence, independent of bed dimensions. Therefore, any reliable approaches for the quantification of IPFs must yield similar results in beds with different diameters and/or heights.

Overshoot surface area in the bed pressure drop profiles: The principle of the presence of an overshoot in the increasing velocity pass of the total bed pressure drop profile of a cohesive bed [5, 46, 50, 57] was employed for a qualitative validation of the results of U_{mf} deviation and DHR approaches. The surface area between the increasing and decreasing velocity pass pressure drop profiles in the peak region is principally impacted by the magnitude of IPFs [57]. In the case of erratic pressure overshoots, which typically happen for very cohesive powders [5], the virtual equivalent peak area must be considered as if there was no cracking in the bed, as illustrated in Figure 4.3 [57]. The values of peak areas at different operating temperatures were normalized by dividing to that at ambient conditions. To qualitatively validate the proposed approaches, the variation of normalized peak area with temperature were then compared with the results obtained from U_{mf} deviation and DHR approaches.

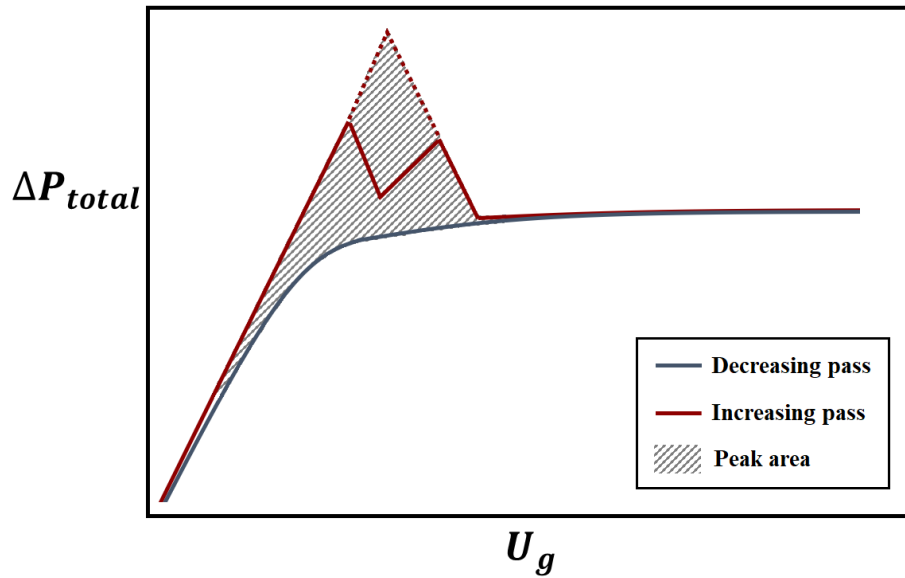


Figure 4.3. Schematic representation of a virtual peak area for an erratic pressure overshoot in the increasing velocity pass of the total bed pressure drop profile at U_{mf} .

4.5 Experimental

4.5.1 Experimental set-up

Fluidization experiments were conducted in an atmospheric pressure, stainless steel fluidized bed capable of operating at 800°C with a fluidization section that has an internal diameter of 5.25 cm and is 500 mm in height. A schematic representation of the adopted rig is presented in Figure 4.4. The fluidizing gas was introduced into the bed through a stainless steel perforated distributor plate having 19 holes arranged in 2 cm triangular pitch. Each distributor nozzle had a hole diameter of 0.5 mm toward the windbox and 3.175 mm toward the fluidization section. The windbox was filled with coarse sand particles, 1-10 mm in size, to help evenly distribute the gas flowing across the unit cross-section and uniformly heat the gas before entering the bed. A blind perforated steel pipe covered with glass wool was employed as a filter in the expansion section of the unit to retain entrained particles in the bed, while imposing the least pressure drop possible on the passing gas.

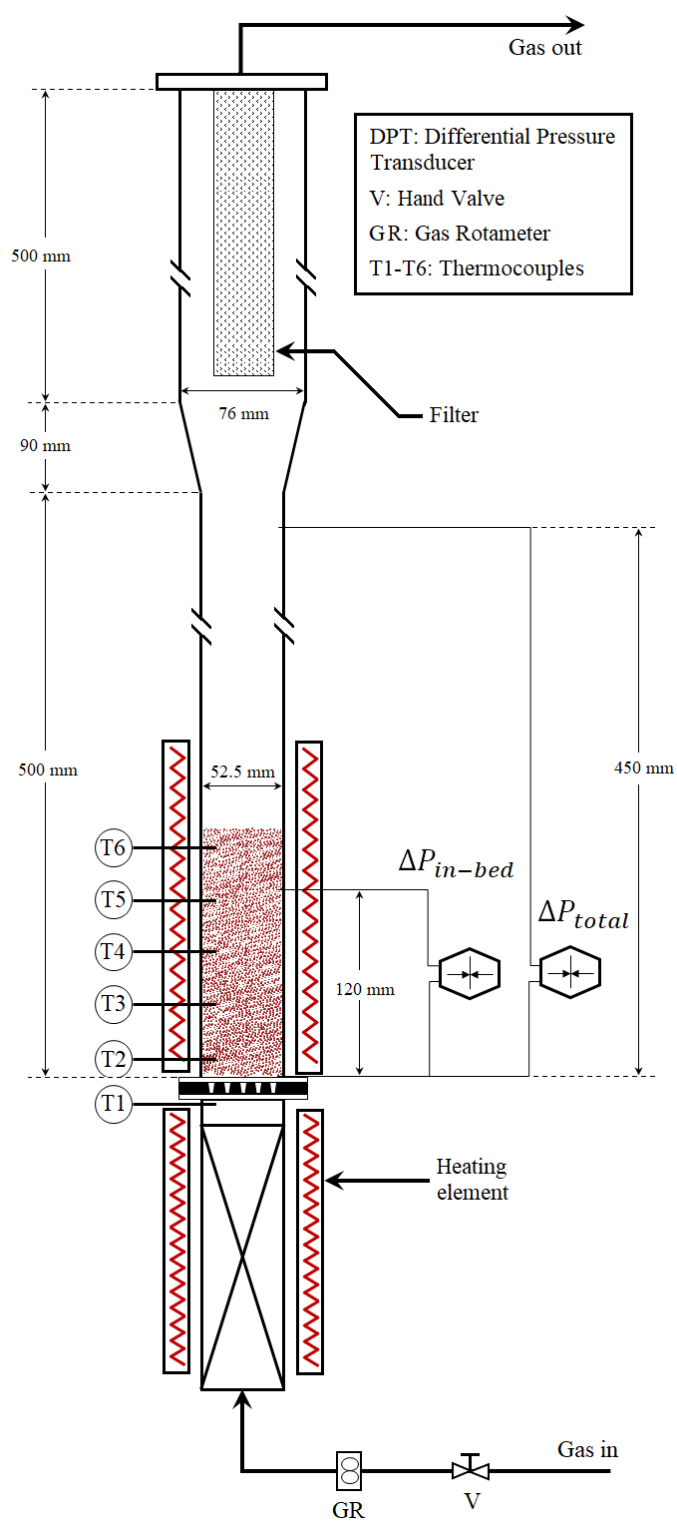


Figure 4.4. Schematic representation of the experimental set-up.

In order to heat the unit to a desired temperature, two sets of heating elements were placed around the rig in the windbox and fluidization section. The first set of heating elements was principally responsible for adjusting the fluidizing gas temperature to the desired value. The second set helped compensate for the heat loss from the unit body, thus keeping the unit surface temperature the same as the inside bed temperature. The unit temperature was monitored with six OMEGA type K thermocouples located at different positions along the axis.

The system was equipped with two calibrated rotameters to accurately adjust the gas flow rate for a wide range of superficial gas velocities. The fluidizing gas flow was measured at room temperature and corrected according to the bed temperature. Two differential pressure transducers were adopted to measure the total bed pressure drop (JUMO, 404304/000-414-415-28-298, 0-100 mbar; 0–45 cm above the distributor plate) and partial in-bed pressure drop (JUMO, 404304/000-412-415-28-298, 0-50 mbar; 0–12 cm above the distributor plate). The loose fixed bed height in all experiments was set to about 15 cm to ensure that the upper leg of the in-bed differential pressure transducer is always placed well below the bed surface and minimally impacted by the splash zone.

4.5.2 Materials

Air and argon were used as fluidizing gases in this study. Their densities at each temperature were estimated based on the ideal gas law as follows:

$$\rho_g = \rho_{g,20^\circ C} \times \left(\frac{293}{T + 273} \right) \quad 4.15$$

where T is the temperature and $\rho_{g,20^\circ C}$ is the gas density at 20°C. The following correlations were adopted to calculate the viscosities of air and argon at different temperatures [65, 66]:

$$\mu_{Air} = 1.46 \times 10^{-6} \frac{(T + 273)^{1.504}}{((T + 273) + 120)} \quad 4.16$$

$$\mu_{Argon} = 10^{-7} \times (44.997 + 0.63892(T + 273) - 0.00012455 (T + 273)^2) \quad 4.17$$

Four solid samples were employed in the experiments: two specimens of glass beads, a fluid cracking catalyst (FCC), and silica sand particles. For simplicity, we refer to these powders with their mean diameters in abbreviated form of GB-67, GB-156, FCC-83, and SS-225, respectively. The physical properties of the adopted powders are given in Table 4.2. Before commencing the hydrodynamic experiments with powders showing typical Geldart group B behaviour, i.e., GB-156 and SS-225, their electrostatic charges were minimized for the sake of comparison with Geldart group A behaviour. To this end, the powders were kept fluidized at 150°C for 3 hrs in the bubbling fluidization regime, while the bed was grounded. The electrostatic charge of the particles will be minimized owing to their enhanced electrical conductivity at this temperature [27, 28], while having enough time to make contact with each other and the grounded bed. The fluidizing gas was, subsequently, stopped and the particles were allowed to gradually cool down to room temperature.

Table 4.2. Characteristics of adopted particles.

Material	Mean particle diameter (μm)	Particle density (kg/m ³)	Sphericity	Geldart group behaviour at ambient conditions	Acronym
Glass beads	67	2595	~1	A	GB-67
FCC	83	1460	~1	A	FCC-83
Glass beads	156	2595	~1	B	GB-156

Silica sand	225	2650	0.74	B	SS-225
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Experiments with GB-67, FCC-83, and SS-225 were conducted at elevated temperature. Sintering of particles can take place at temperatures higher than the minimum sintering temperature. If this happens, it causes the level of IPFs and fluidization behaviour to change over time due to the time dependency of the sintering process [25]. Hence, the experiments must be carried out at temperatures lower than this temperature to avoid misleading results. The minimum sintering temperature of these particles was measured by a NETZSCH DIL-402 dilatometer in this study. Consequently, the experiments with GB-67 powders were carried out up to 200°C, while the maximum temperature in experiments with other particles was limited to 650°C.

4.5.3 Measurement procedures

4.5.3.1 U_{mf} , ε_0 , and ε_{mf}

The minimum fluidization velocity was determined from the total bed pressure drop profile in the decreasing velocity pass according to Richardson's classical method [67]. U_{mf} was defined as the superficial gas velocity corresponding to the intersection of the pressure drop line for the fixed bed state with the relatively constant pressure drop at the fully fluidized state. For each fluidization test and at each gas velocity, the average bed height, L_{ave} , was calculated assuming a linear pressure drop profile along the bed height:

$$L_{ave} = \frac{\Delta P_{total}}{\Delta P_{in-bed}} l \quad 4.18$$

where ΔP_{total} and ΔP_{in-bed} are the total and in-bed pressure drops, and l is the distance between two legs of the in-bed differential pressure transducer (see Figure 4.4). Then, the average bed voidage at each superficial gas velocity, ε_{ave} , can be calculated as:

$$\varepsilon_{ave} = 1 - \frac{m_s/\rho_p}{AL_{ave}} \quad 4.19$$

Upon plotting ε_{ave} profile against U_g , the bed voidage at U_{mf} in the decreasing velocity pass was considered as ε_{mf} . The bed voidage for particles with the typical behaviour of Geldart group A gradually decreases as U_g is gently decreased from U_{mf} to zero and reaches a final value equal to the fixed bed voidage in the increasing velocity pass (see Figure 4.7). This value was, thus, reported as ε_0 and is equal to the loose bulk voidage.

4.5.3.2 Water volume fraction in water-solid system

When the gaseous phase is replaced by water in a particulate bed, no material bridge can form between particles. Electrostatic interaction between two particles is inversely proportional to the electric permittivity of the environment based on Coulomb's law [13, 68]. Therefore, electrostatic and van der Waals forces are negligible due to the high relative electric permittivity of water (≈ 80 for water in comparison with ≈ 1 for air at 20°C). Therefore, a minimal level of IPFs acts on particles when they are in water. The water volume fraction in a water-solid system, ε_w , thus, denotes the bulk voidage in the virtual absence of interparticle forces.

A special device was used for the measurement of ε_w . A porous plate was installed at the bottom of a graduated cylinder with a diameter identical to the adopted fluidized bed's internal diameter. Dry powders were gently poured into the graduated cylinder in their loose bulk state. To measure ε_w with minimal destruction to the particulate assembly, water was gently introduced into the graduated cylinder from the bottom porous plate. The particulate assembly, hence, maintained its structure when water filled the gaps between particles. ε_w was then calculated by applying Eq. 4.19, while the bed height was measured by visual observation of the particulate assembly's height in the graduated cylinder.

4.6 Results and discussion

4.6.1 Proof of concept for U_{mf} deviation approach

Since U_{mf} is one of the most critical fluidization characteristics of powders, many correlations have been proposed to predict it [69]. For instance, the Carman–Kozeny equation [55] can be expressed as:

$$U_{mf,pred} = \frac{(\phi_s d_p)^2}{C} \frac{\rho_p - \rho_g}{\mu_g} g\left(\frac{\varepsilon_{mf}^3}{1 - \varepsilon_{mf}}\right) \quad 4.20$$

where the value of constant C is 150 based on the Ergun equation [54]. However, it depends on the tortuosity of the flow path [70]. Researchers reported C values around 180 for relatively coarse spherical particles [50]. In our case, C values of 173, 188, and 163 for SS-225, GB-67, and FCC-83, respectively, provided the best match with the corresponding experimental U_{mf} at ambient conditions. The Carman-Kozeny equation is valid in the laminar flow regime, where the viscous effect dominates over inertia at the minimum fluidization condition, i.e., when $Re_{p,mf} < 20$, if IPFs are insignificant. $Re_{p,mf}$ were in the range of 0.01 to 0.61 in our experiments over the explored operating conditions.

A comparison between the experimental U_{mf} at different temperatures and that predicted by the Carman-Kozeny equation for SS-225, GB-67, and FCC-83, when fluidized by air and argon, are presented in Figure 4.5. It shows that $U_{mf,exp}$ for SS-225 monotonously decreased with temperature, while it experienced a trend inversion around 100°C and increased with temperature at higher temperatures for GB-67 and FCC-83. The variations of experimental U_{mf} with temperature in this study are in broad agreement with the results reported in the literature for similar powders [29, 56, 61, 71, 72]. At low $Re_{p,mf}$, where HDFs are dominated by the viscous effect, U_{mf} is inversely proportional to the gas viscosity if the effect of IPFs is not pronounced [10, 30]. Under this condition, U_{mf} is expected to decrease as the gas viscosity increases with temperature. Figure 4.5a shows that the operating temperature, in the range explored, had minimal

effect on the level of IPFs between SS-225 particles since the temperature variation of the corresponding $U_{mf,exp}$ follows the trend predicted by the Carman-Kozeny equation. This can be attributed to the intermediate size and high melting point of these particles. However, $U_{mf,exp}$ for GB-67 and FCC-83 deviates from the prediction of the Carman-Kozeny equation at high temperatures, while it can be adequately predicted by about 100°C. Refer to Figure 4.5b and Figure 4.5c, the Carman-Kozeny equation is capable of accounting for the effect of gas properties on U_{mf} as it accurately predicts U_{mf} of GB-67 and FCC-83 with both air and argon at ambient conditions with the same constant C . Since the variation of HDFs and its impact on U_{mf} is considered in the Carman-Kozeny equation, the observed deviation of $U_{mf,exp}$ from $U_{mf,pred}$ at high temperatures can be attributed to an enhanced level of cohesive IPFs. This concept was, therefore, applied in Section 4.3.1 to develop the U_{mf} deviation approach for the quantification of thermally induced IPFs.

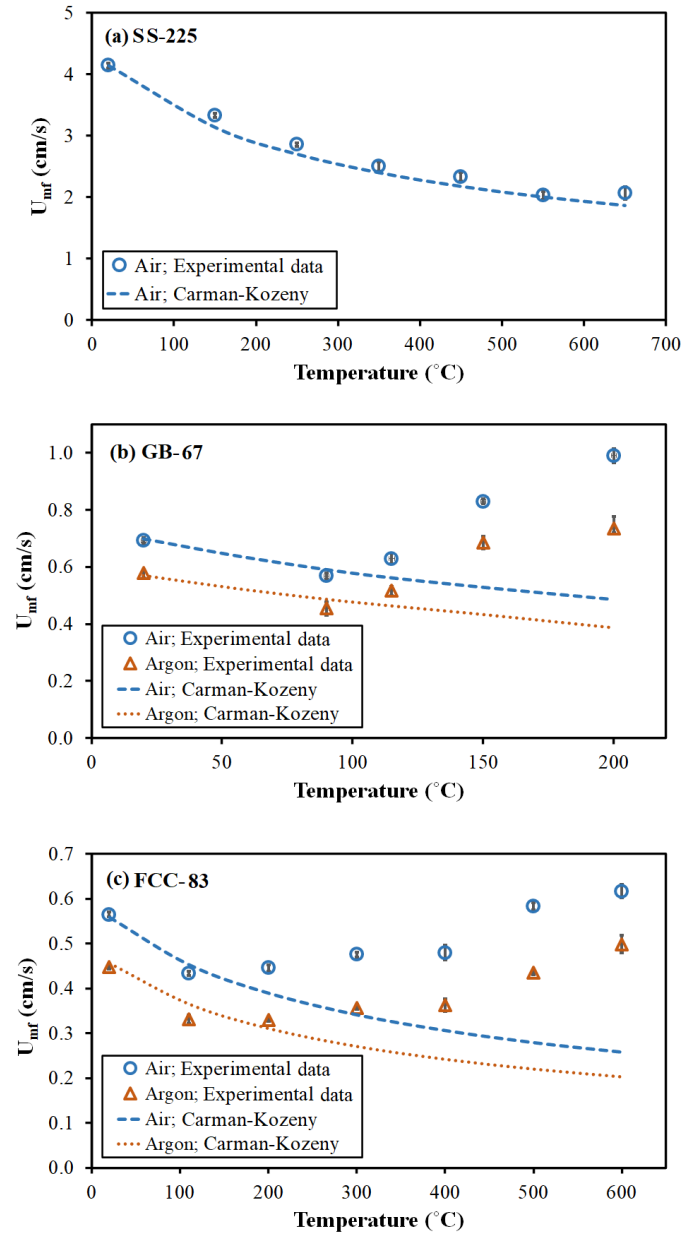


Figure 4.5. A comparison of $U_{mf,exp}$ and $U_{mf,pred}$ for a) SS-225, b) GB-67, and c) FCC-83 (ϵ_{mf} is considered at its corresponding values at ambient conditions in the Carman-Kozeny equation).

4.6.2 Proof of concept for DHR approach

The ε_0 and ε_{mf} were measured for all particles adopted in this study at ambient conditions and compared with the corresponding ε_w , as presented in Figure 4.6. Since ε_w corresponds to the minimum level of IPFs in a particulate assembly (see Section 4.5.3.2), the extent of the difference between ε_w and ε_0 highlights the significance of IPFs in the particulate bed in the loose bulk state. Following this remark, results in Figure 4.6 show the presence of an appreciable level of IPFs in the case of GB-67 and FCC-83 (showing typical behaviour of Geldart group A) in contrast to GB-156 and SS-225 (showing typical behaviour of Geldart group B) for which the levels of IPFs were relatively negligible.

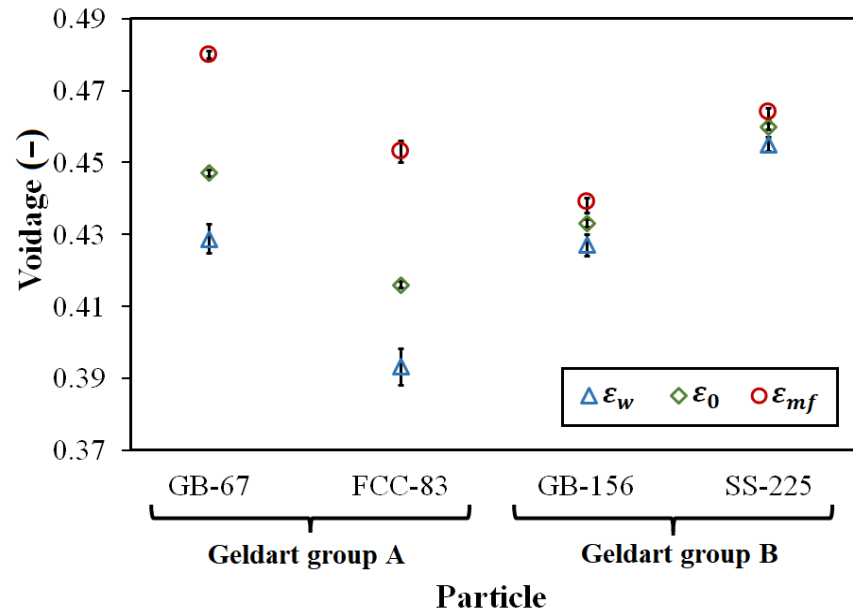


Figure 4.6. ε_0 , ε_{mf} , and ε_w for particles adopted in this study with typical behaviours of Geldart group A and B at ambient conditions.

The representative total bed pressure drop and corresponding voidage profiles of particles with typical behaviours of Geldart group A (GB-67) and B (GB-156) are presented in Figure 4.7. The total bed pressure drop profile of the former shows a discernible overshoot at the U_{mf} in the

increasing velocity pass, which corresponds to the difference between ε_0 and ε_{mf} (Figure 4.7a and Figure 4.7b). However, for the latter, there is no overshoot in the increasing velocity pass pressure drop profile as the corresponding ε_0 and ε_{mf} are practically identical (Figure 4.7c and Figure 4.7d).

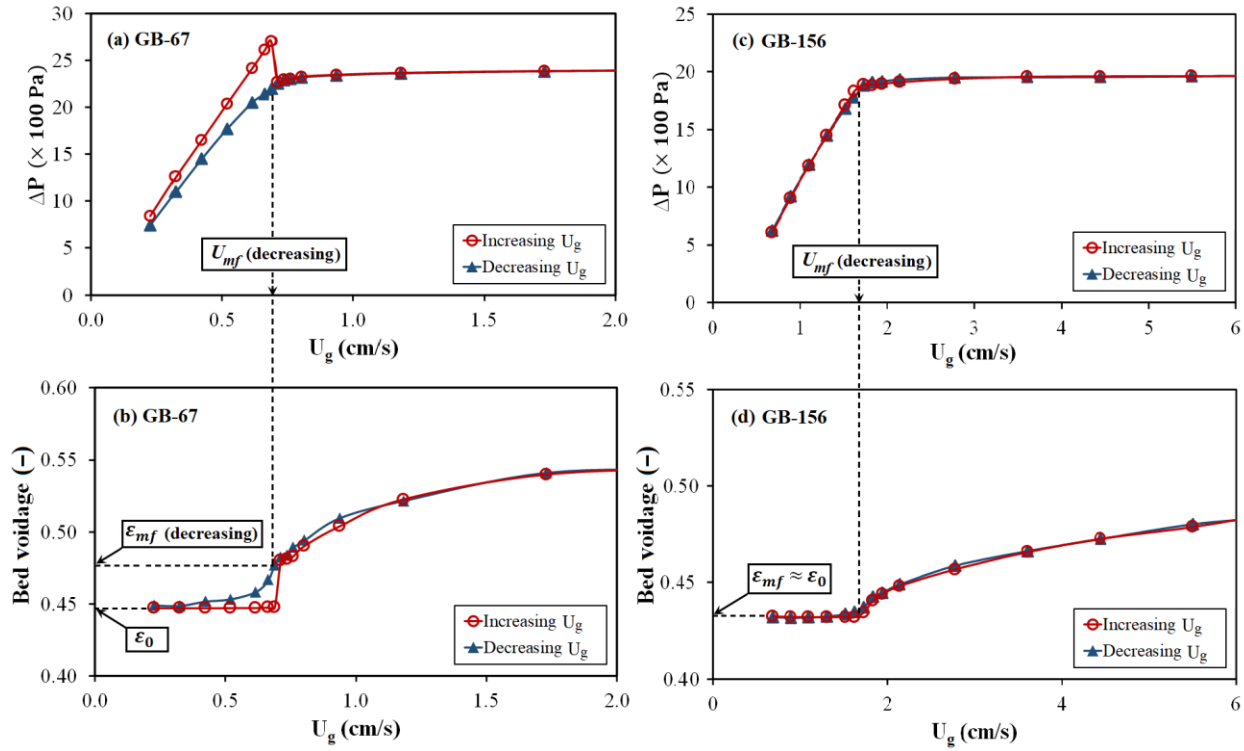


Figure 4.7. Representative bed pressure drop and corresponding voidage profiles of particles with typical behaviours of Geldart group A (GB-67) and B (GB-156) at ambient conditions.

To better understand the underlying phenomenon for the difference between ε_0 and ε_{mf} and highlight the effect of IPFs, the temperature variations of ε_0 and ε_{mf} for GB-67 and FCC-83 were studied. Although air and argon were adopted as the fluidizing gases in the current series of experiments, the measured ε_0 and ε_{mf} were independent of the gas type. The results, presented in Figure 4.8, show that ε_0 and ε_{mf} for both particles slightly decreased up to about 100°C after which they increased with temperature. The observed generally increasing trends of ε_0 and ε_{mf}

with temperature in this study are in accordance with the reported experimental trends in the literature [21, 29, 56, 61, 72-74]. Similar minimums around 100°C in the ε_{mf} -temperature profile have also been reported in the literature [56, 72, 73]. Several studies have shown that both ε_0 and ε_{mf} increase as the level of IPFs grows [5, 29, 74]. Therefore, for a specific particulate assembly, a larger ε_0 and/or ε_{mf} can be interpreted as a higher level of IPFs in the system. Integrating this remark with the results presented in Figure 4.8 suggest that the level of IPFs in beds of GB-67 and FCC-83 decreased up to around 100°C and, subsequently, increased with temperature. This conclusion is in accordance with the variation of corresponding normalized peak areas with temperature (see Figure 4.10).

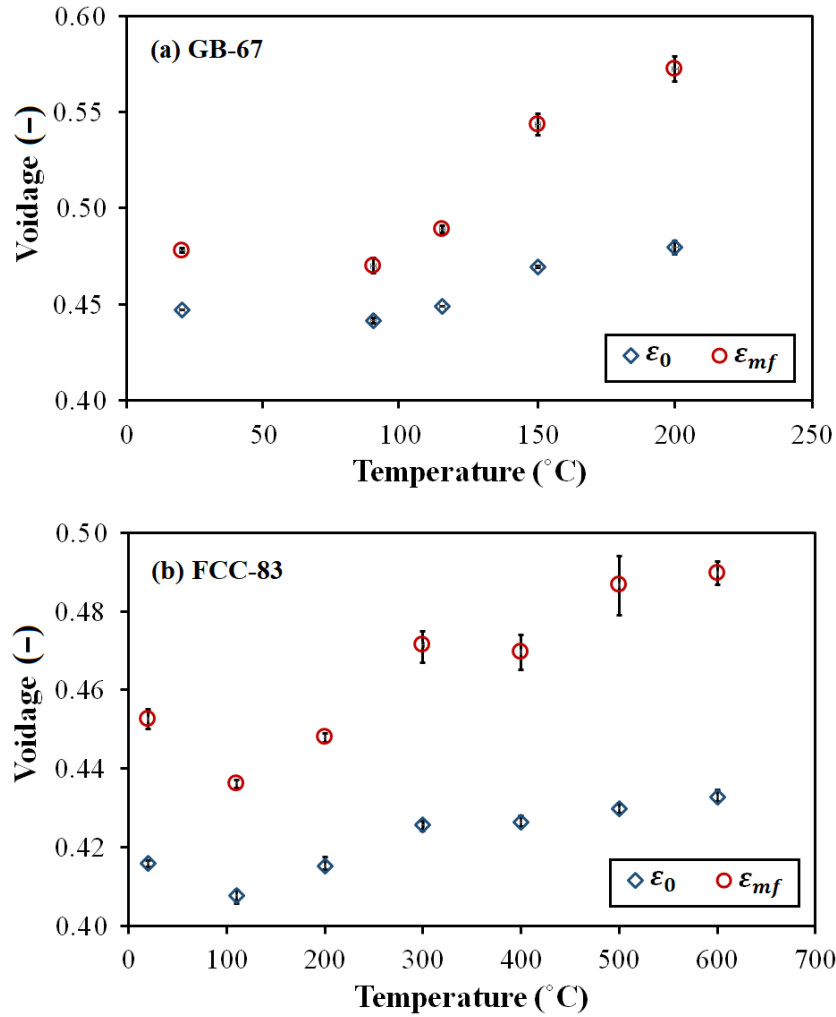


Figure 4.8. Variations of ϵ_0 and ϵ_{mf} with temperature for a) GB-67 and b) FCC-83 (air and/or argon as the fluidizing gas).

The comparison of ϵ_0 and ϵ_{mf} in Figure 4.8 reveals that not only the values of ϵ_0 and ϵ_{mf} but also the difference between them were impacted by the level of IPFs. The values of ϵ_{mf}/ϵ_0 at different temperatures, plotted against the corresponding ϵ_0 in Figure 4.9, show that DHR linearly increased with ϵ_0 for both particles. Extrapolating these trendlines to the point where $\epsilon_{mf}/\epsilon_0 = 1$, leads to $\epsilon_{mf} = \epsilon_0 = 0.425$ for GB-67 and $\epsilon_{mf} = \epsilon_0 = 0.383$ for FCC-83. These values are very close to the ϵ_w of these particles, $\epsilon_w = 0.429 \pm 0.004$ for GB-67 and 0.392 ± 0.005 for FCC-83. These

findings together with the similarity of ε_{mf} , ε_0 , and ε_w for particles with negligible IPFs (see Figure 4.6) indicate that DHR can relate the level IPFs and bed behaviour at incipient fluidization conditions.

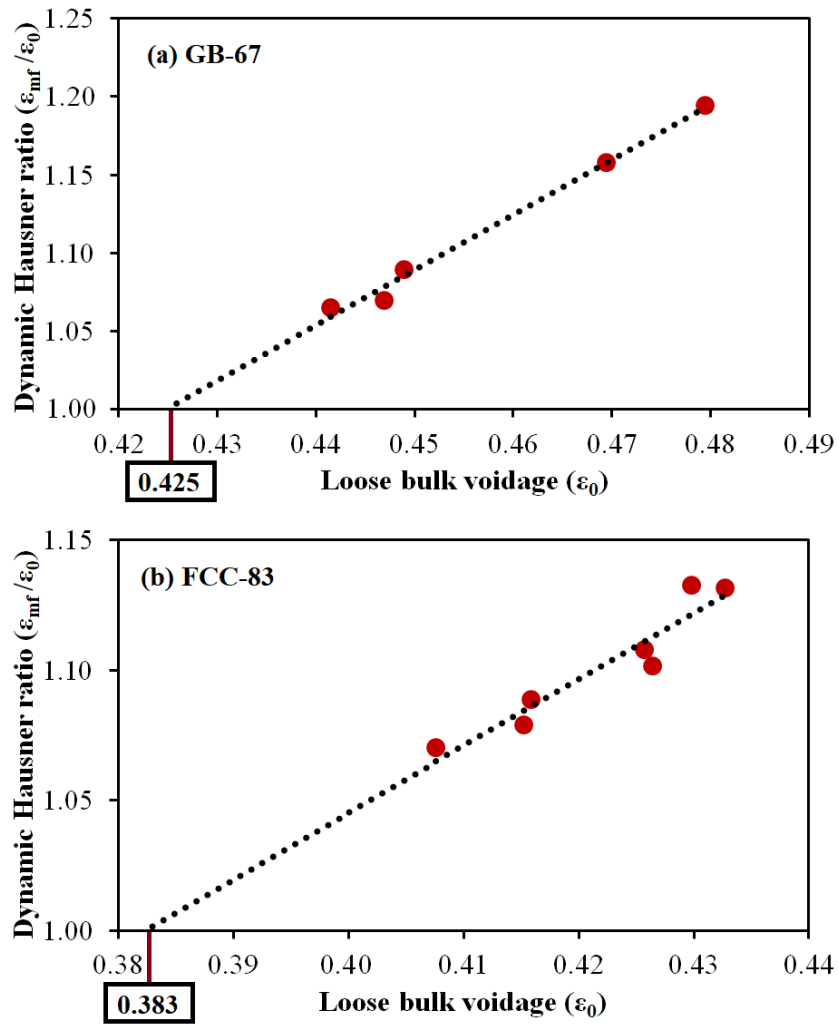


Figure 4.9. Variation of dynamic Hausner ratio with the loose bulk voidage for a) GB-67 and b) FCC-83.

An approximate analysis of the distance between adjacent particles in a particulate assembly, as presented in the Supplementary Material, suggests that the distance at minimum fluidization conditions (ε_{mf}) is discernibly larger than at the fixed bed state (ε_0). This, in turn, implies that the magnitude of IPFs at minimum fluidization conditions is appreciably smaller than that at the fixed bed state for particles showing typical behaviour of Geldart group A. The experimental data and supporting discussions presented in this section reinforce the idea presented in Section 4.3.2 for the development of the DHR approach and Eq. 4.14 to quantify the level of IPFs in a gas-solid fluidized bed.

4.6.3 Performances of U_{mf} deviation and DHR approaches

Eqs. 4.5 and 4.14 as the outcomes of U_{mf} deviation and DHR approaches were applied to quantify the magnitudes of IPFs and their thermal variation for GB-67 and FCC-83. Results are presented in Figure 4.10 and compared with the temperature variation of the normalized peak area. Both approaches highlighted a slight decrease in the level of IPFs up to about 100°C and an appreciable increase at higher temperatures for the adopted samples. The initial decline in the level of IPFs can be attributed to the dominant decrease in the magnitude of electrostatic forces [26, 27]. With a further increase in the operating temperature, van der Waals forces and their thermal variation likely dominate the magnitude of IPFs as it can appreciably grow with the rising temperature [10, 22, 68]. In addition, Figure 4.10 demonstrates that the results of both approaches change with temperature in line with the trends observed for the normalized peak area, which also confirm the minimum IPFs at about 100°C.

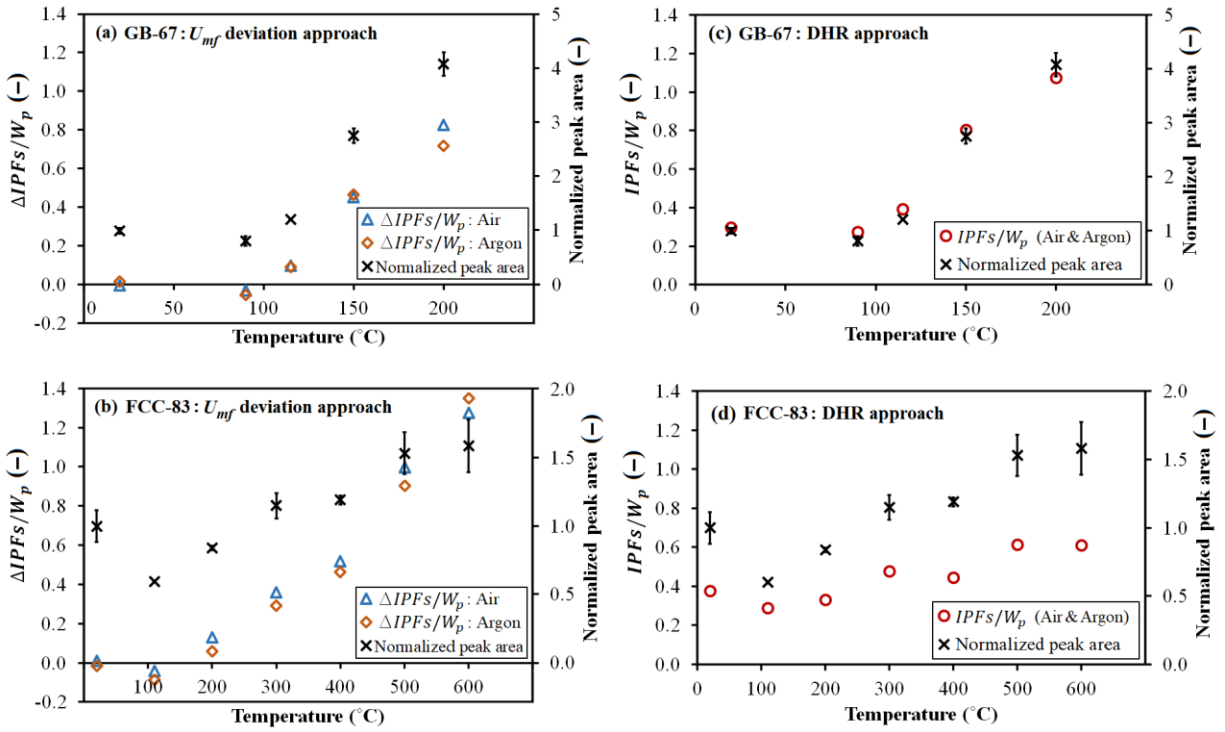


Figure 4.10. $\Delta IPFs/W_p$ quantified by Eq. 4.5 for a) GB-67 and b) FCC-83, and $IPFs/W_p$ quantified by Eq. 4.14 for c) GB-67 and d) FCC-83 in comparison with the thermal variation of the normalized peak area.

GB-67 and FCC-83 showed typical behaviour of Geldart group A over the explored experimental conditions. The magnitude of IPFs and the particle's weight are in the same order for particles showing this class of behaviour [12, 19]. In accordance with these remarks, quantified values of $IPFs/W_p$ using the DHR approach vary in the range of 0.27-1.07 for GB-67 and 0.29-0.61 for FCC-83 (Figure 4.10c and Figure 4.10d). In addition, $\varepsilon_{mf}/\varepsilon_0$ is close to one for particles showing the typical behaviour of Geldart group B (Figure 4.6), which leads to the quantification of the small values of $IPFs/W_p$ for these particles, as expected. Similarly, if one adds $\Delta IPFs/W_p$ quantified by the U_{mf} deviation approach to $IPFs/W_p$ at ambient conditions, the obtained results will be in the expected range. To further support the obtained values by the proposed approaches, the van der Waals forces between two adjacent GB-67 particles at ambient conditions were estimated by

the Hamaker [75] and Rumpf [76] models employing the available data in the literature [77]. Although the estimation involves a high degree of uncertainty, the calculated values of $IPFs/W_p$ by the Hamaker and Rumpf models (= 0.36 and 0.58, respectively) are comparable with the result of the DHR approach (= 0.3). The estimated values were obtained under the most plausible conditions, i.e., the particle radius was replaced by the asperity radius in the Hamaker model, and the separation distance was equated to 0.4 nm [13, 25].

Due to minimal gas adsorption on the surface of particles under the conditions explored in this study, IPFs are expected to be independent of the gas type used in this study. The application of the U_{mf} deviation approach resulted in the quantification of similar $\Delta IPFs/W_p$ at a given temperature when employing air and argon as the fluidizing gas (Figure 4.10a and Figure 4.10b). In addition, similar $IPFs/W_p$ were calculated by the DHR approach in the experiments with air and argon (Figure 4.10c and Figure 4.10d) since the gas type did not affect ε_0 and ε_{mf} in the first place.

The U_{mf} , ε_0 , and ε_{mf} are intrinsic characteristics of particles and, basically, independent of the bed dimensions. Consequently, the results of Eqs. 4.5 and 4.14 will not essentially vary with the bed diameter and/or height. This is in agreement with the authors' unpublished data showing that the DHR for GB-67 and FCC-83 at ambient conditions in two beds with internal diameters of 5.25 and 15.25 cm and different bed heights from 15 to 40 cm are comparable within an experimental error of 2.5%. In addition, a maximum 7% difference in the U_{mf} of each powder was observed by changing the bed internal diameter and/or height.

According to the results presented in Figure 4.10 and the above discussions, the proposed approaches were verified and qualitatively validated. They offer a number of advantages as listed below:

- (i) The proposed approaches are in-situ and inspired by the phenomena taking place in a gas-solid fluidized bed.

- (ii) While most developed approaches for the quantification of IPFs have practical limitations under extreme operating conditions, the proposed approaches in this study are capable of being conveniently employed under such conditions.
- (iii) No fluidization assisting method is required for the proposed approaches. In addition, the measurements of required parameters, i.e., U_{mf} , ε_0 , and ε_{mf} , for these approaches are relatively simple.
- (iv) Not only Eq. 4.14, as the outcome of the DHR approach, is capable of quantifying the magnitude of IPFs in a gas-solid fluidized bed, but it has also led to a more comprehensive understanding of bed behaviour at minimum fluidization conditions and the underlying reason for the difference between ε_0 and ε_{mf} .

The U_{mf} deviation and DHR approaches can offer discernible opportunities for improving the performance of commercial processes employing gas-solid fluidized beds and further research and development in fluidization technology. They can be implemented for the development of reliable hydrodynamic correlations that are capable of considering the effects of IPFs, particularly under extreme conditions. The availability of these correlations, which can adequately predict the hydrodynamics of gas-solid fluidized beds under extreme conditions, is critical for their successful design, scale-up, operation, and performance evaluation. The developed approaches in this study can also assist in the implementation of a more realistic computational fluid dynamics-DEM (CFD-DEM) simulation of fluidized beds in the presence of IPFs.

4.6.4 Limitations and integration of the proposed approaches

While the DHR approach quantifies the absolute values of IPFs, the U_{mf} deviation approach can only quantify the difference between IPFs at a given temperature and that at ambient conditions. Hence, the absolute value of $IPFs/W_p$ at a given temperature can be alternatively calculated by adding $\Delta IPFs/W_p$ at the temperature of interest quantified by the U_{mf} deviation approach to $IPFs/W_p$ at ambient conditions quantified by the DHR approach:

$$\frac{IPFs}{W_p}(at\ T) = \frac{IPFs}{W_p}(at\ ambient) + \frac{\Delta IPFs}{W_p} \quad 4.21$$

The values of $IPFs/W_p$ quantified by Eq. 4.14 and those calculated by Eq. 4.21, resulting from the integration of the proposed approaches (the sum of $\frac{IPFs}{W_p}$ at ambient conditions and $\frac{\Delta IPFs}{W_p}$ calculated by Eq. 4.5), were compared in Figure 4.11a and Figure 4.11b for GB-67 and FCC-83, respectively. The effective agglomerate diameter at minimum fluidization conditions, $d_{agg,mf}$, was estimated from the Ergun equation, as presented below, and illustrated in Figure 4.11c and Figure 4.11d.

$$d_{agg,mf} = \sqrt{\frac{150U_{mf}\mu_g L_{mf}(1 - \varepsilon_{mf})^2}{\varepsilon_{mf}^3 \Delta P_{U_{mf}, \varepsilon_{mf}}}} \quad 4.22$$

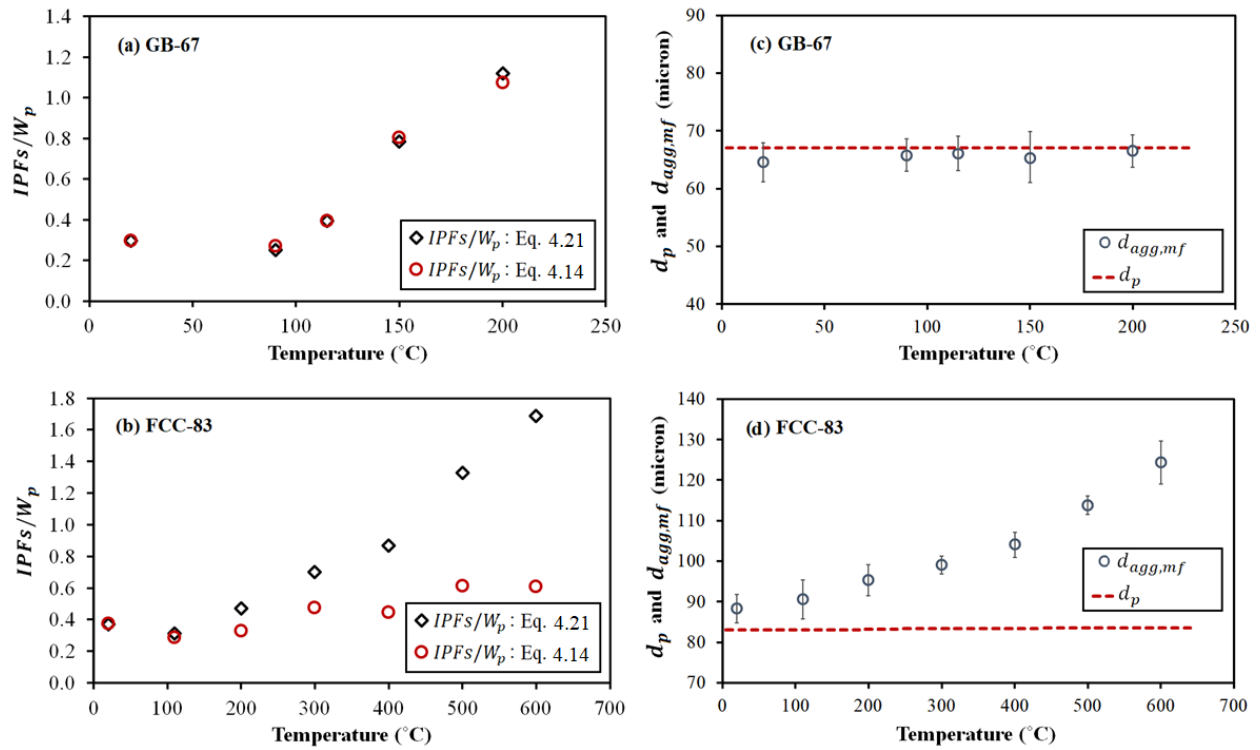


Figure 4.11. Comparison of the results of U_{mf} deviation and DHR approaches and the effect of agglomerating fluidization.

Figure 4.11a shows that the values of $IPFs/W_p$ quantified by Eqs. 4.21 and 4.14 closely match for GB-67 indicating that the DHR and U_{mf} deviation approaches support each other in this case. The results presented in Figure 4.11c reveal that GB-67 particles were likely fluidized in the form of individual particles, in accordance with the assumption adopted for the development of the DHR approach. FCC-83 particles, however, formed agglomerates, which were larger at higher temperatures (Figure 4.11d). The deviation from the adopted assumption, based on which Eq. 4.14 was developed, yields an under-estimation of IPFs by the DHR approach, in particular at elevated temperatures. Hence, the DHR approach must be generalized to transcend its limitation and extend its capability to quantify IPFs under agglomerating fluidization conditions. This is critical for particles with typical Geldart group C behaviour, which are usually fluidized in the form of large

agglomerates of their primary particles [13, 14]. Nevertheless, Eq. 4.14 gives a reasonable estimation of IPFs when the agglomerate size is close to the particle size, for example at ambient conditions for FCC-83. Therefore, Eq. 4.21 can be employed to obtain a realistic estimation of IPFs at higher temperatures for FCC-83. Adopting such an integrated strategy offers a way to apply the DHR and U_{mf} deviation approaches more extensively, particularly when large agglomerates form at elevated temperatures, while the agglomerate size is close to the primary particle size at ambient conditions. It is also advantageous from the practical perspective as the measurement of U_{mf} is more convenient than that of ε_0 and ε_{mf} , particularly at elevated temperatures.

4.7 Conclusion

This work presents two simple and indirect approaches for the quantification of IPFs and its variation with temperature in a gas-solid fluidized bed. The U_{mf} deviation approach is established based on the difference between the experimental U_{mf} and that predicted by an appropriate solely hydrodynamic correlation. The DHR approach is based on the difference between ε_0 and ε_{mf} , as the basis for the appearance of an overshoot at U_{mf} in the bed pressure drop profile. A model was, subsequently, developed to quantify IPFs as a function of DHR, which was experimentally shown to be closely related to the level of IPFs.

The proposed approaches were applied to quantify IPFs for GB-67 and FCC-83 at operating temperatures ranging from ambient to 600°C. The key results of this study are as follows:

- In broad agreement with findings reported in the literature for powders similar to those adopted in this study, the magnitudes of IPFs estimated by the proposed approaches were in the same order as the particle's weight.
- In accordance with expectations and owing to negligible gas adsorption on the surface of particles under the conditions explored in this study, the gas type had no effect on the quantified IPFs by the proposed approaches.

- The proposed approaches are basically independent of the reactor dimension and/or the bed height.
- The quantified IPFs by the proposed approaches were qualitatively validated by the temperature variation of a normalized peak area.
- Under the conditions explored in this study, a slight decrease in the level of IPFs up to about 100°C, which was followed by an appreciable increase with temperature, was observed. The first part of this drift could be attributed to a dominant decrease in the level of electrostatic forces, whereas the increase in the level of van der Waals forces likely determines the growth part of the observed evolution.

The U_{mf} deviation and DHR approaches can help develop reliable hydrodynamic correlations that simultaneously consider the effects of IPFs and HDFs. They also assist in the implementation of comprehensive CFD-DEM models for the simulation of fluidized beds in the presence of IPFs. Despite the promising performances achieved by the proposed approaches, the U_{mf} deviation approach can only quantify the magnitude of thermally induced IPFs, and not its absolute values. However, the DHR approach can quantify the absolute values of IPFs if particles are individually fluidized. Extending the capability of the DHR approach to also be applicable to agglomerating fluidization could be the subject of future studies. Nevertheless, integrating the proposed approaches in this study helps estimate the magnitude of IPFs at various operating conditions under both agglomerating and individual particle fluidization.

Acknowledgements

The authors would like to acknowledge TOTAL American Services, Inc. for their financial support of this research study.

4.8 Supplementary Material: Insight into the difference between ε_0 and ε_{mf} based on particle-particle distance

Several studies have suggested that asperity sizes of particles, rather than their particle radii/harmonic radius, must be adopted for a realistic estimation of typical IPFs, e.g., van der Waals and electrostatic forces [13, 25]. This is due to a greater contribution of molecules at the frontier surfaces to the level of van der Waals and electrostatic forces between adjacent particles [68]. Hence, to find a phenomenological justification for the difference between ε_0 and ε_{mf} for particles showing typical behaviour of Geldart group A, an analysis of the average distance between surfaces of adjacent particles in a particulate assembly, a_s , with respect to the bed voidage, ε , is desirable.

To obtain a relationship between a_s and ε , let us consider a particulate assembly of identical spheres positioned in a cubic configuration as shown in Figure 4.12. The number of particles in a row of length L in every direction, $N_{p,i}$, can be calculated as:

$$N_{p,i} = \frac{L}{a_{s,cub} + d_p} \quad 4.23$$

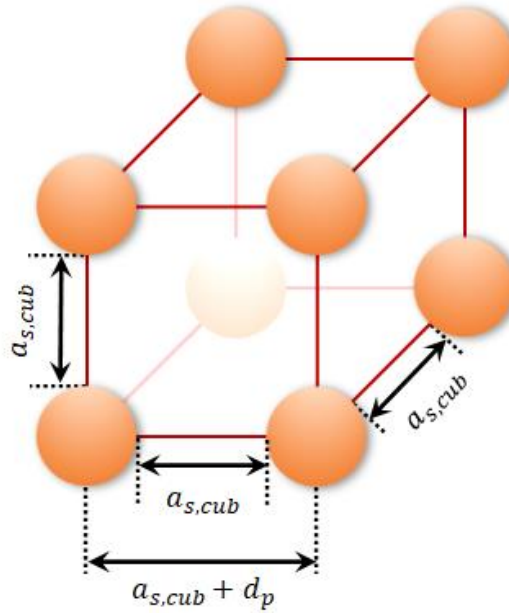


Figure 4.12. Schematic representation of particles positioned in a body-centered cubic configuration.

where $a_{s,cub}$ is the distance between the surfaces of adjacent particles in the cubic configuration. Hence, the total number of particles in a cubic bed of dimensions L is equal to:

$$N_p = N_{p,i}^3 = \left(\frac{L}{a_{s,cub} + d_p} \right)^3 \quad 4.24$$

By multiplying N_p to the volume of a particle, V_p , the total volume occupied by this number of particles, $V_{p,total}$, can be calculated as:

$$V_{p,total} = N_p V_p = \left(\frac{L}{a_{s,cub} + d_p} \right)^3 \left(\frac{\pi d_p^3}{6} \right) \quad 4.25$$

Therefore, the bed voidage can be calculated as:

$$\varepsilon = \frac{L^3 - V_{p,total}}{L^3} = 1 - \frac{\left(\frac{L}{a_{s,cub} + d_p}\right)^3 \left(\frac{\pi d_p^3}{6}\right)}{L^3} \quad 4.26$$

By rearranging Eq. 4.26 for $a_{s,cub}$, we obtain:

$$a_{s,cub} = d_p \left[\left(\frac{\pi}{6(1 - \varepsilon)} \right)^{\frac{1}{3}} - 1 \right] \quad 4.27$$

Following the same procedure as described above, the following equations were derived for a_s for triangular and semi cubic-triangular configurations:

$$a_{s,tri} = d_p \left[\left(\frac{2\pi}{9(1 - \varepsilon)} \right)^{\frac{1}{3}} - 1 \right] \quad (\text{Triangular configuration}) \quad 4.28$$

$$a_{s,cub-tri} = d_p \left[\left(\frac{\pi}{3\sqrt{3}(1 - \varepsilon)} \right)^{\frac{1}{3}} - 1 \right] \quad (\text{Semi cubic-triangular configuration}) \quad 4.29$$

where $a_{s,tri}$ and $a_{s,cub-tri}$ are the distances between surfaces of adjacent particles when they are structured in triangular and semi cubic-triangular configurations, respectively. Calculated values of $a_{s,cub}$, $a_{s,tri}$, and $a_{s,cub-tri}$ by Eqs. 4.27-4.29 for an assembly of 67-micron particles are plotted in Figure 4.13. Since the loose bulk voidage increases with IPFs, we can assume that the particle-particle distance is close to zero when virtually no IPFs act on the particles, i.e., in water. Therefore, by knowing ε_w , a_s for the GB-67 system can be estimated by the dashed curve in Figure 4.13 as an interpolation of calculated $a_{s,cub}$ and $a_{s,cub-tri}$. The ratio of a_s at minimum fluidization conditions (ε_{mf}) to that at the loose bulk state (ε_0) was quantified at around 3.0 on the dashed curve at all temperatures explored in this study. This ratio was around 2.4 for FCC-83 particles.

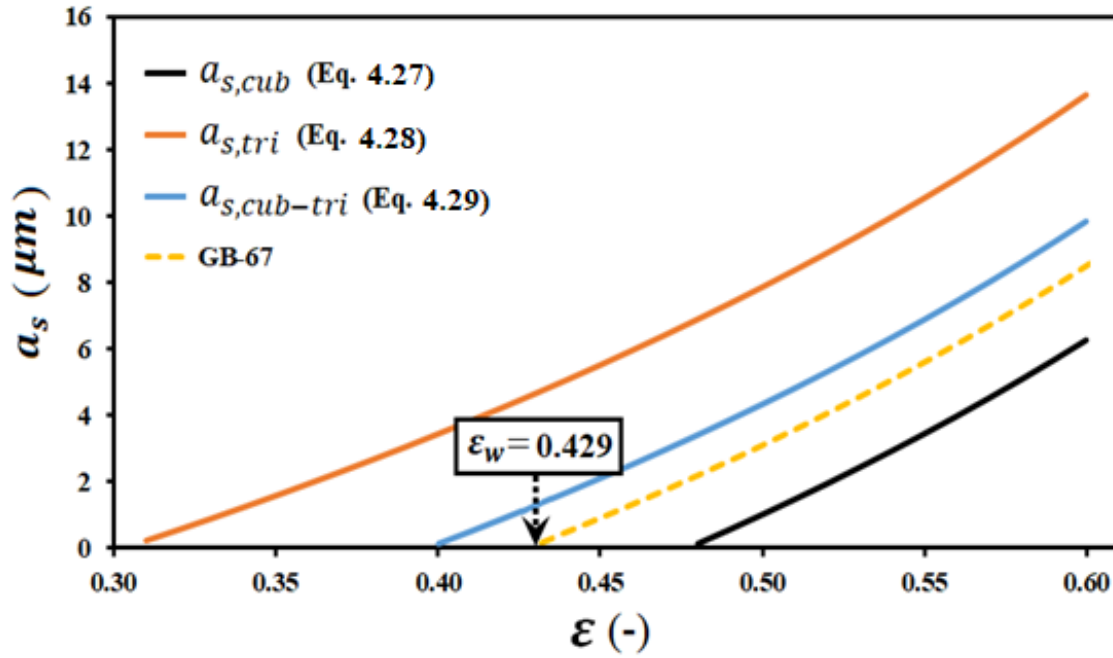


Figure 4.13. $a_{s,cub}$, $a_{s,tri}$, and $a_{s,cub-tri}$ for an assembly of 67-micron spherical particles and prediction of a_s for GB-67 against bed voidage.

The van der Waals and electrostatic forces are inversely proportional to the square of particle-particle distance [13, 25]. The relationships for the estimation of material bridge forces are more complicated. However, if liquid bridge forces are present, they are also expected to substantially decrease by increasing a_s [25]. Knowing the ratio of a_s at ϵ_{mf} to that at ϵ_0 and assuming that the magnitude of IPFs is proportional to $\left(\frac{1}{a_s^2}\right)$, the ratio of IPFs acting on particles at ϵ_{mf} to that at ϵ_0 were calculated at around 0.11 and 0.17 for GB-67 and FCC-83, respectively. Although this analysis is not accurate, we can still ensure that the ratio of IPFs at minimum fluidization conditions to that at the loose bulk state is small for particles behaving like Geldart group A. This formed a basis for the development of the DHR approach in Section 4.3.2. These values are also in accordance with the calculated fraction of IPFs acting on particles at minimum fluidization conditions ($1 - F_{fp,mf} = 0.16$ and 0.23 for GB-67 and FCC-83, respectively) based on the fluidization quality index at U_{mf} explained in assumptions (v) and (vi) in Section 4.3.2.1.

4.9 Nomenclature

Acronyms

AFM	atomic force microscopy
CFD-DEM	computational fluid dynamics coupled with discrete element method
DEM	discrete element method
DHR	dynamic Hausner ratio
FCC	fluid cracking catalyst
FCC-83	83-micron fluid cracking catalyst
GB-67	67-micron glass beads
GB-156	156-micron glass beads
HDFs	hydrodynamic forces
HR	Hausner ratio
IPFs	interparticle forces
SS-225	225-micron silica sand

Symbols

a_s	distance between surfaces of adjacent particles (m), see Supplementary material
$a_{s,cub}$	distance between surfaces of adjacent particles in cubic configuration (m), see Supplementary material

$a_{s,tri}$	distance between surfaces of adjacent particles in triangular configuration (m), see Supplementary material
$a_{s,cub-tri}$	distance between surfaces of adjacent particles in semi cubic-triangular configuration (m), see Supplementary material
A	bed cross sectional area (m ²)
C	constant in the Carman–Kozeny equation (-)
$d_{agg,mf}$	effective agglomerate diameter at minimum fluidization conditions (m)
d_p	particle diameter (m)
$F_{fp,mf}$	fraction of fluidized particles at minimum fluidization conditions (-)
g	gravitational acceleration (m/s ²)
$IPFs$	interparticle forces acting on each particle at loose bulk state (N)
$IPFs_{total}$	total interparticle forces that are overcome when the bed voidage increases from ε_0 to ε_{mf} (N)
l	distance between two legs of the in-bed differential pressure transducer (m)
L	length (m), see Supplementary material
L_{ave}	average bed height (m)
L_{mf}	bed height at minimum fluidization conditions (m)
L_0	bed height at loose bulk state (m)
m_s	bed inventory (kg)
N_p	total number of particles in the bed (-)
$N_{p,i}$	number of particles in a row of length L (-), see Supplementary material
$Re_{p,mf}$	particle Reynolds number at minimum fluidization conditions (-)

T	temperature (°C)
U_g	superficial gas velocity (m/s)
U_{mf}	minimum fluidization velocity (m/s)
$U_{mf,exp}$	experimental minimum fluidization velocity (m/s)
$U_{mf,pred}$	minimum fluidization velocity predicted by an appropriate correlation (m/s)
V_p	volume of a particle (m ³), see Supplementary material
$V_{p,total}$	total volume of particles in a particulate assembly (m ³), see Supplementary material
W_p	weight of a single particle (N)

Greek letters

$\Delta IPFs$	thermally induced IPFs acting on each particle (N)
$\Delta IPFs_{total}$	total thermally induced IPFs in the bed at a given temperature (N)
ΔP_{eq}	equivalent bed pressure drop (Pa)
ΔP_{in-bed}	in-bed pressure drop (Pa)
ΔP_{total}	total bed pressure drop (Pa)
$\Delta P_{U_{mf}, \varepsilon_{mf}}$	bed pressure drop at minimum fluidization velocity and the voidage of ε_{mf} (Pa)
$\Delta P_{U_{mf}, \varepsilon_0}$	bed pressure drop at minimum fluidization velocity and the voidage of ε_0 (Pa)
ε	voidage (-)
ε_0	loose bulk voidage (-)
ε_{ave}	average bed voidage (-)

ε_{mf}	minimum fluidization voidage (-)
ε_w	water volume fraction in water-solid system (-)
μ_{Air}	air viscosity (Pa. s)
μ_{Argon}	argon viscosity (Pa. s)
μ_g	gas viscosity (Pa. s)
ρ_g	gas density (kg/m ³)
$\rho_{g,20^\circ C}$	gas density at 20°C (kg/m ³)
ρ_p	particle density (kg/m ³)
ϕ_s	particle shape factor (-)

CHAPTER 5 ARTICLE 2 : QUANTIFICATION OF INTERPARTICLE FORCES IN GAS-AGGLOMERATED PARTICLES FLUIDIZED BEDS

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Published on 20 June 2023, Chemical Engineering Journal

DOI: <https://doi.org/10.1016/j.cej.2023.144181>

Highlights:

- Quantification of IPFs by the ratio of experimental and theoretical values of U_{mf} .
- Generalized dynamic Hausner ratio equation correlating IPFs and agglomerate size.
- Correspondence between Geldart group behaviours and magnitude of IPFs.

5.1 Abstract

Interparticle forces (IPFs) dominate the fluidization of fine and ultrafine particles and are responsible for the formation of agglomerates. Two equivalent equations are proposed in this study for the quantification of IPFs in a gas-solid fluidized bed. The first is the generalized U_{mf} deviation equation correlating the magnitude of IPFs to the ratio of experimental and theoretical minimum fluidization velocities. The second is the generalized form of the Dynamic Hausner Ratio (DHR) equation, which was developed based on the appearance of an overshoot in the bed pressure drop profile at U_{mf} . The generalized DHR equation relates the magnitude of IPFs to the agglomerate

size in addition to the DHR, i.e., the ratio of minimum fluidization and loose bulk voidages. The proposed equations can be employed under various conditions, including ambient and elevated operating conditions, as well as individual and agglomerated particles fluidization. They help quantify the resultant IPFs acting on particles irrespective of the types of these forces. The quantified IPFs by the proposed equations were verified and qualitatively validated with different principles. The results indicated that different fluidization behaviours correspond to varying magnitudes of IPFs. The opportunities and limitations of the proposed equations are further discussed.

Keywords: Gas-solid fluidized bed, Hydrodynamics, Interparticle forces, Particle agglomeration, Quantification.

5.2 Introduction

The hydrodynamics of a gas-solid fluidized bed is determined by gravitational, hydrodynamic forces (HDFs), and interparticle forces (IPFs) acting on particles and their relative importance [1-3]. For instance, research has indicated that particular ratios of IPFs and the weight of a particle W_p corresponds to the transition between Geldart groups [4] C, A and B [1, 5, 6]. While the magnitude of IPFs is typically negligible in comparison with HDFs and W_p for coarse particles [1, 7], it governs the fluidization behaviour of fine and ultrafine powders showing Geldart group C behaviour [1, 8, 9].

Research studies have shown that upon enhancing IPFs to a moderate level, the bed voidage, including the loose bulk, minimum fluidization, and emulsion phase voidages, increases [7, 10-12]. Accordingly, the fluidizing gas is more prone to pass through the bed in the emulsion phase in the bubbling fluidization regime [12-14]. These modifications in the bed hydrodynamics can enhance the overall performance of a catalytic gas-solid fluidized bed reactor [12, 13]. A substantial increase in the level of IPFs can alter its beneficial impacts to deleterious effects through the formation of particle agglomerates and channeling [6, 8, 9] and may lead to bed defluidization under extreme conditions [15, 16].

The influence of IPFs on the bed hydrodynamics and its overall performance underlines the need for a reliable approach to quantify these forces. A review of the available methods for the qualitative/quantitative evaluation of IPFs was reported in a recent publication by the group [7]. Most of these approaches are qualitative and/or ex-situ, i.e., must be accomplished outside of a gas-solid fluidized bed, or need an assisting fluidization method. Hence, they are hardly applicable to the conditions of a particulate assembly in a fluidized bed. Accordingly, we developed the “dynamic Hausner ratio (DHR)” and “ U_{mf} deviation” (U_{mf} is the minimum fluidization velocity) approaches for an in-situ quantification of the magnitude of IPFs and their thermal variation in a gas-solid fluidized bed [7]. The U_{mf} deviation approach was developed to quantify the growth in the level of IPFs with temperature, i.e., the difference between the magnitude of IPFs at a given temperature and that at ambient conditions. It was established based on the difference between the experimental minimum fluidization velocity at a given temperature $U_{mf,exp}$ and the prediction of Carman-Kozeny [17] equation $U_{mf,pred}$, which is based purely on hydrodynamic principles. The U_{mf} deviation approach is incapable of quantifying the absolute magnitude of IPFs and its value at ambient conditions. The DHR approach, however, can quantify the absolute magnitude of IPFs if particles are individually fluidized, but underestimates the magnitude of these forces in agglomerating fluidization. This approach was developed based on the appearance of an overshoot in the bed pressure drop profile at U_{mf} resulting from the difference between the fixed bed voidage ε_0 and the minimum fluidization voidage ε_{mf} . The DHR approach's limitation in quantifying the magnitude of IPFs in agglomerating fluidization is particularly important for fine and ultrafine powders. These powders are typically fluidized in the form of agglomerates of primary particles, which could be considerably larger than their building blocks [6, 8, 9].

In this paper, we present generalized forms of the U_{mf} deviation and DHR approaches to extend their capabilities in quantifying the magnitude of IPFs in agglomerating gas-solid fluidized beds. The generalized equations help distinguish between fractions of IPFs that are overcome and continue to act during bed fluidization. It will also be shown that the non-generalized DHR equation can be equally employed in agglomerating fluidization if ε_{mf} is modified to account for the fraction of IPFs that continue to act on particles after fluidization.

5.3 Model development

5.3.1 Background

In a recent study by the authors [7], two approaches were proposed for the estimation of the magnitude of IPFs and its variation with temperature in a gas-solid fluidized bed. Assumptions adopted for the development of the non-generalized U_{mf} deviation and DHR approaches [7] include unisize spherical particles and viscous flow regime, where the particle Reynolds number at minimum fluidization $Re_{p,mf}$ is lower than about 20 [18, 19]. These assumptions are equally applied to their generalized forms, which are presented in this work. The non-generalized DHR equation for estimating the absolute magnitude IPFs acting on each particle at a given operating condition is as follows:

$$\frac{IPFs}{W_p} = \left(\frac{\varepsilon_{mf}}{\varepsilon_0} \right)^3 \frac{1 - \varepsilon_0}{1 - \varepsilon_{mf}} - 1 \quad 5.1$$

To avoid any confusion about the notation adopted throughout this document, note that all parameters except those with a subscript indicating the corresponding value at ambient conditions, e.g., $\varepsilon_{mf,amb}$, refer to their values at conditions of interest.

Eq. 5.1 was derived assuming that the total magnitude of IPFs is proportional to the overshoot pressure drop at U_{mf} resulting from the difference between ε_0 and ε_{mf} . The adopted assumption is valid only in the fluidization of particles with a relatively modest level of IPFs, where virtually no IPFs act on particles after being fluidized [7]. In agglomerating fluidization, however, it is invalid since a significant fraction of IPFs will act on primary particles within agglomerates even after fluidization. This leads to an underestimation of the magnitude of IPFs by Eq. 5.2 under agglomerating fluidization conditions.

The U_{mf} deviation approach was developed to quantify the magnitude of thermally induced IPFs, i.e., the difference between the magnitude of IPFs at a given temperature and that at ambient

conditions, $\Delta IPFs$. The result of this approach, as presented in the following equation, correlates $\Delta IPFs$ to the difference between $U_{mf,exp}$ and $U_{mf,pred}$:

$$\frac{\Delta IPFs}{W_p} = \frac{C_{Erg}}{d_p^2} \frac{1 - \varepsilon_{mf,amb}}{\varepsilon_{mf,amb}^3} \frac{\mu_g}{\rho_p g} (U_{mf,exp} - U_{mf,pred}) \quad 5.2$$

where d_p and ρ_p are the particle size and its density, μ_g is the gas viscosity, and C_{Erg} and g are the constant in the Ergun [20] equation ($= 150$) and the gravitational acceleration, respectively. The $U_{mf,pred}$ in Eq. 5.2 was estimated by a hydrodynamic based correlation, e.g., Carman–Kozeny equation, that adequately considers the effect of gas properties, while neglecting the impact of IPFs. The variations of ε_0 and ε_{mf} with temperature are solely attributed to the change in the level of IPFs [7, 11] and thus should not be considered in the calculation of $U_{mf,pred}$. The ε_{mf} in the Carman-Kozeny equation was, hence, considered as its value at ambient conditions for the calculation of $U_{mf,pred}$:

$$U_{mf,pred}|_{\varepsilon_{mf,amb}} = \frac{d_p^2}{C_{C-K}} \frac{\rho_p - \rho_g}{\mu_g} g \left(\frac{\varepsilon_{mf,amb}^3}{1 - \varepsilon_{mf,amb}} \right) \quad 5.3$$

where ρ_g and C_{C-K} are the gas density and the constant of the Carman-Kozeny equation, respectively. The latter can be expressed as:

$$C_{C-K} = \frac{C_{Erg}}{F_{fp,mf}} \quad 5.4$$

where $F_{fp,mf}$ is the fraction of fluidized particles at U_{mf} [7] (see Supplementary Material and Section 5.3.2 for further details).

Although Eq. 5.2 can be employed for the estimation of $\Delta IPFs$ in both individual particle and agglomerating fluidization conditions, it is incapable of quantifying the absolute magnitude of IPFs, including its value at ambient conditions. The limitations of the non-generalized DHR and U_{mf} deviation approaches overlap if agglomerates appreciably greater than the primary particles form at ambient conditions. This is usually the case for fine and ultrafine particles as a result of

their high level of IPFs compared to HDFs and/or W_p . The generalized forms of the U_{mf} deviation and DHR approaches to extend their capabilities to these circumstances will be presented in Sections 5.3.2 and 5.3.3, respectively.

5.3.2 Generalized U_{mf} deviation approach

For the development of Eq. 5.2, Soleimani et al. [7] divided the force corresponding to the difference between $U_{mf,exp}$ and $U_{mf,pred}$ by the total weight of particles in the bed, implicitly assuming that all particles are fluidized at minimum fluidization conditions. In the DHR approach, however, the force required to overcome IPFs was divided by the weight of the fluidized particles at U_{mf} rather than the total weight of the particles [7]. The fraction of fluidized particles at minimum fluidization $F_{fp,mf}$ is assumed to be equal to the ratio of the experimental bed pressure drop at U_{mf} to the equivalent pressure drop, i.e., the weight of the bed per its cross-sectional area. The latter strategy must be applied to the U_{mf} deviation approach as it is more plausible. Accordingly, dividing the force corresponding to the difference between $U_{mf,exp}$ and $U_{mf,pred}$ by the weight of the fluidized particles at U_{mf} , gives:

$$\frac{\Delta IPFs}{W_p} = \frac{C_{Erg}}{F_{fp,mf} d_p^2} \frac{1 - \varepsilon_{mf,amb}}{\varepsilon_{mf,amb}^3} \frac{\mu_g}{(\rho_p - \rho_g)g} (U_{mf,exp} - U_{mf,pred}) \quad 5.5$$

By substituting Eqs. 5.3 and 5.4 in Eq. 5.5, and since $\rho_p - \rho_g \approx \rho_p$ in a gas-solid system, we obtain:

$$\frac{\Delta IPFs}{W_p} = \frac{U_{mf,exp}}{U_{mf,pred}|_{\varepsilon_{mf,amb}}} - 1 \quad 5.6$$

Eq. 5.6 can be regarded as the modified U_{mf} deviation equation yet not in its generalized form to be employed for quantification of the absolute magnitude of IPFs. It quantifies the magnitude of thermally induced IPFs acting on each particle at a given temperature with respect to the ambient

conditions. It remains incapable of quantifying the absolute values of IPFs when $U_{mf,pred}$ is estimated by Eq. 5.3.

Eq. 5.6 was developed based on the incapability of purely hydrodynamic correlations, like Carman-Kozeny equation, in adequate estimation of $U_{mf,exp}$ under an enhanced level of IPFs [2, 21]. However, research has shown that the Carman-Kozeny equation is able to adequately predict the $U_{mf,exp}$ if the experimental value of ε_{mf} – rather than ε_0 or $\varepsilon_{mf,amb}$ – is employed in this equation [11, 22]. This can only be achieved for particles with the typical behaviour of Geldart group A in the case of individual particle fluidization [7, 11, 22]. Hence, experimental minimum fluidization velocity under these circumstances $U_{mf,exp,ind}$ can be adequately estimated by $U_{mf,pred}|_{\varepsilon_{mf}}$ (see Supplementary Material):

$$U_{mf,exp,ind} \approx U_{mf,pred}|_{\varepsilon_{mf}} = \frac{d_p^2}{C_{C-K}} \frac{\rho_p - \rho_g}{\mu_g} g \left(\frac{\varepsilon_{mf}^3}{1 - \varepsilon_{mf}} \right) \quad 5.7$$

Substituting Eqs. 5.3 and 5.7 in Eq. 5.6 results in:

$$\left. \frac{\Delta IPFs}{W_p} \right|_{ind} = \left(\frac{\varepsilon_{mf}}{\varepsilon_{mf,amb}} \right)^3 \frac{1 - \varepsilon_{mf,amb}}{1 - \varepsilon_{mf}} - 1 \quad 5.8$$

Eq. 5.8 closely resembles the non-generalized DHR equation, Eq. 5.1, which helps quantify the absolute magnitude of IPFs. The only difference between these two equations is the substitution of $\varepsilon_{mf,amb}$ in Eq. 5.8 for ε_0 in Eq. 5.1. The comparison of Eqs. 5.1 and 5.8 reveals that the non-generalized DHR and U_{mf} deviation approaches can be considered as different formats of Eq. 5.6. Eq. 5.6 is equivalent to Eq. 5.8 and quantifies the magnitude of thermally induced IPFs when $\varepsilon_{mf,amb}$ is employed in the Carman-Kozeny equation for the estimation of $U_{mf,pred}$. The absolute magnitude of IPFs can be quantified by Eq. 5.6 if ε_0 is substituted for $\varepsilon_{mf,amb}$ in the calculation of $U_{mf,pred}$. Thus, Δ must be omitted from Eq. 5.6 in the latter case:

$$\frac{IPFs}{W_p} = \frac{U_{mf,exp}}{U_{mf,pred}|_{\varepsilon_0}} - 1 \quad 5.9$$

Under individual particle fluidization, Eq. 5.9 will be equivalent to the non-generalized DHR equation, i.e., Eq. 5.1.

$U_{mf,pred}$ can be estimated at $\varepsilon_{mf,amb}$ (adopted in Eq. 5.6) or ε_0 (adopted in Eq. 5.9), whereas $U_{mf,exp}$ is inherently measured at ε_{mf} . The equivalence of the DHR and U_{mf} deviation approaches shows that they originated from a similar phenomenon. The DHR equation was developed based on the overshoot pressure drop corresponding to the difference between ε_0 and ε_{mf} . Hence, one can also think of Eqs. 5.6 and 5.9 as the estimation of IPFs by the pressure drop corresponding to the difference between ε_{mf} and $\varepsilon_{mf,amb}$, and that between ε_0 and ε_{mf} , respectively.

The equivalence of Eqs. 5.9 and 5.1 under individual particle fluidization explains the similarity of the quantified IPFs with the non-generalized U_{mf} deviation and DHR approaches for 67-micron glass beads in our recent publication [7]. In contrast, the formation of agglomerates in the fluidization of FCC particles led to an underestimation of the magnitude of IPFs by the non-generalized DHR equation (Eq. 5.1) for these powders [7]. The generalized form of Eq. 5.1, equivalent to Eq. 5.9, will be derived in Section 5.3.3.

Baerns [23] was the first to point out the close relationship between IPFs and the fluidizability index, i.e., the ratio of theoretical and experimental minimum fluidization velocities. Eqs. 5.9 and 5.6 demonstrate this relationship and highlight the importance of employing an appropriate voidage in the Carman- Kozeny equation for the estimation of $U_{mf,pred}$. Experimental and predicted minimum fluidization velocities are the only parameters that are required to be known in Eq. 5.9. One can, therefore, take advantage of the simplicity of this equation to quantify the magnitude of IPFs in a wide range of conditions, from ambient to elevated operating conditions, in individual particle and agglomerating fluidization.

5.3.3 Generalized DHR approach

The experimental value of the minimum fluidization velocity in agglomerating fluidization $U_{mf,exp,agg}$ cannot be adequately estimated by the Carman-Kozeny equation even after substituting ε_{mf} for ε_0 . In this case, one must replace d_p by $d_{agg,mf}$, i.e., the effective agglomerate size at minimum fluidization conditions, in addition to using the experimental value of ε_{mf} in the Carman-Kozeny equation (see Supplementary Material):

$$U_{mf,exp,agg} \approx U_{mf,pred}|_{\varepsilon_{mf} \& d_{agg,mf}} = \frac{d_{agg,mf}^2}{C_{C-K}} \frac{\rho_p - \rho_g}{\mu_g} g\left(\frac{\varepsilon_{mf}^3}{1 - \varepsilon_{mf}}\right) \quad 5.10$$

As alluded to earlier, to quantify the absolute magnitude of IPFs by Eq. 5.9, $U_{mf,pred}$ must be calculated with ε_0 as the bed voidage in the Carman-Kozeny equation:

$$U_{mf,pred}|_{\varepsilon_0} = \frac{d_p^2}{C_{C-K}} \frac{\rho_p - \rho_g}{\mu_g} g\left(\frac{\varepsilon_0^3}{1 - \varepsilon_0}\right) \quad 5.11$$

Substituting Eqs. 5.10 and 5.11 in Eq. 5.9 yields:

$$\frac{IPFs}{W_p} = \left(\frac{d_{agg,mf}}{d_p}\right)^2 \left(\frac{\varepsilon_{mf}}{\varepsilon_0}\right)^3 \frac{1 - \varepsilon_0}{1 - \varepsilon_{mf}} - 1 \quad 5.12$$

Eq. 5.12 is the generalized form of the DHR equation and is simplified to Eq. 5.1 when particles fluidize individually. To quantify the magnitude of IPFs, Eq. 5.12 requires the measurement of agglomerate size, which might be challenging under certain circumstances. This equation, however, leads to a more comprehensive understanding of the fluidization behaviour of cohesive particles and the underlying relationship between IPFs and the formation of agglomerates. The difference between ε_{mf} and ε_0 , which is mainly manifested in the $\frac{\varepsilon_{mf}}{\varepsilon_0}$ ratio in Eq. 5.12, represents IPFs that must be overcome for the bed fluidization. In contrast, the formation of agglomerates, expressed by $\frac{d_{agg,mf}}{d_p}$ in Eq. 5.12, signifies the contribution of the remaining IPFs that act on

particles after fluidization. A summary of the generalized and non-generalized U_{mf} deviation and DHR equations and their application conditions is presented in Table. 1.

Table 5.1. Summary of the U_{mf} deviation and DHR equations and their application conditions.

Name	Equation	Remarks
Non-generalized U_{mf} deviation equation (modified version) (Eq. 5.6)	$\frac{\Delta IPFs}{W_p} = \frac{U_{mf,exp}}{U_{mf,pred} _{\varepsilon_{mf,amb}}} - 1$	* Quantifies the difference between IPFs at a given operating conditions and that at ambient conditions.
Generalized U_{mf} deviation equation (Eq. 5.9)	$\frac{IPFs}{W_p} = \frac{U_{mf,exp}}{U_{mf,pred} _{\varepsilon_0}} - 1$	* Equivalent to Eq. 5.12. * Quantifies the absolute magnitude of IPFs at any operating conditions.
Non-generalized DHR equation (Eq. 5.1)	$\frac{IPFs}{W_p} = \left(\frac{\varepsilon_{mf}}{\varepsilon_0}\right)^3 \frac{1 - \varepsilon_0}{1 - \varepsilon_{mf}} - 1$	* Quantifies the absolute magnitude of IPFs at any operating conditions but only under individual particle fluidization. * Underestimates IPFs in agglomerating fluidization.
Generalized DHR equation (Eq. 5.12)	$\frac{IPFs}{W_p} = \left(\frac{d_{agg,mf}}{d_p}\right)^2 \left(\frac{\varepsilon_{mf}}{\varepsilon_0}\right)^3 \frac{1 - \varepsilon_0}{1 - \varepsilon_{mf}} - 1$	* Equivalent to Eq. 5.9. * Quantifies the absolute magnitude of IPFs at any operating conditions.

The variation of $\frac{IPFs}{W_p}$ with $\frac{\varepsilon_{mf}}{\varepsilon_0}$ and $\frac{d_{agg,mf}}{d_p}$ based on Eq. 5.12 at a representative $\varepsilon_{mf}(= 0.6)$ is illustrated in Figure 5.1. Different regions in Figure 5.1 correspond to particles with different Geldart group behaviours, from coarse group B to fine group C particles, as specified in the figure. In this regard, four scenarios can be considered:

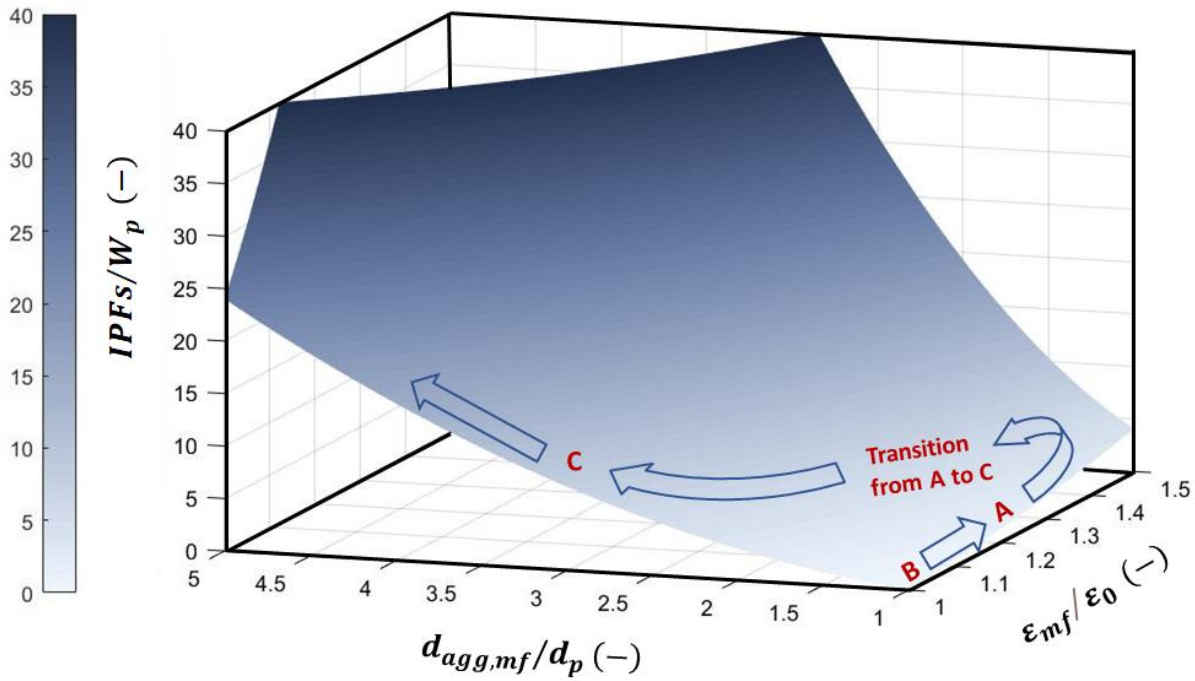


Figure 5.1. Variation of $\frac{IPFs}{W_p}$ with $\frac{\varepsilon_{mf}}{\varepsilon_0}$ and $\frac{d_{agg,mf}}{d_p}$ based on Eq. 5.12 at $\varepsilon_{mf} = 0.6$.

- 1) If $\frac{IPFs}{W_p} \approx 0$, particles fluidize individually at a ε_{mf} very close to the corresponding ε_0 . These are typical fluidization characteristics of intermediate-sized and coarse particles with Geldart group B behaviour and correspond to the bottom right origin in Figure 5.1 ($\frac{\varepsilon_{mf}}{\varepsilon_0} \approx 1$, $\frac{d_{agg,mf}}{d_p} \approx 1$, and $\frac{IPFs}{W_p} \approx 0$).

- 2) When $0 < \frac{IPFs}{W_p} < \sim 1$, ε_{mf} deviates from ε_0 and results in the appearance of an overshoot pressure drop at U_{mf} while particles essentially fluidize individually. This is the typical behaviour of coarser particles with Geldart group A behaviour and corresponds to the axis of $\frac{\varepsilon_{mf}}{\varepsilon_0}$ in Figure 5.1 ($\frac{\varepsilon_{mf}}{\varepsilon_0} > 1$ and $\frac{d_{agg,mf}}{d_p} \approx 1$). In this case, the magnitude of IPFs can be quantified by the non-generalized DHR equation, Eq. 5.1.
- 3) An additional increase in the level of IPFs when it remains in the same order as W_p ($\sim 1 < \frac{IPFs}{W_p} < \sim 10$) not only causes ε_{mf} to be greater than ε_0 but also leads to the formation of agglomerates slightly greater than the primary particle size ($1 < \frac{d_{agg,mf}}{d_p} < \sim 3$). This behaviour can be considered the transition from Geldart group A to C behaviour and is observed for smaller particles with Geldart group A behaviour. Similar behaviour is also reported for intermediate-sized and coarse particles fluidized under an enhanced level of IPFs, for example, if the IPFs are artificially amplified by particle coating [13] or wetting [24] or at elevated temperatures [7].
- 4) Channeling is one of the distinguished fluidization characteristics of powders with Geldart group C behaviour [6, 8]. It implies that the bed does not expand at U_{mf} and, hence, ε_{mf} is very similar to ε_0 for particles in this class of behaviour. The order of magnitude of IPFs is greater than W_p for these particles ($\frac{IPFs}{W_p} > \sim 10$). The behaviour of these particles falls on the axis of $d_{agg,mf}/d_p$ in Figure 5.1 ($\frac{\varepsilon_{mf}}{\varepsilon_0} \approx 1$, and $\frac{d_{agg,mf}}{d_p} > \sim 3$). Under these circumstances, the magnitude of IPFs is mainly reflected in $d_{agg,mf}/d_p$ in Eq. 5.12, which reduces to:

$$\frac{IPFs}{W_p} \approx \left(\frac{d_{agg,mf}}{d_p} \right)^2 - 1 \quad 5.13$$

Accordingly, the $d_{agg,mf}$ can be estimated if the magnitude of IPFs is known:

$$\frac{d_{agg,mf}}{d_p} \approx \sqrt{\frac{IPFs}{W_p} + 1} = \sqrt{\frac{U_{mf,exp}}{U_{mf,pred}|_{\varepsilon_0}}} \quad 5.14$$

IPFs are influenced by many factors, including material properties (such as surface properties, conductivity, and the Hamaker constant), operating conditions, humidity, and external fields. The two equivalent equations presented in this study, i.e., Eqs. 5.9 and 5.12, help back-calculate the magnitude of the resultant IPFs acting on each particle from their ultimate effects on the fluidization parameters. These parameters include $U_{mf,exp}$ in Eq. 5.9, and $\frac{d_{agg,mf}}{d_p}$ and $\frac{\varepsilon_{mf}}{\varepsilon_0}$ in Eq. 5.12. For instance, any growth in the level of IPFs, regardless of its source, causes $U_{mf,exp}$ to increase [2] and will be reflected in Eq. 5.9. This feature makes the proposed equations independent of the source and type of IPFs.

To cross-check and qualitatively validate the quantified IPFs by the generalized U_{mf} deviation and DHR approaches, i.e., Eqs. 5.9 and 5.12, an alternative model is presented in Section 5.3.4. We called this model the modified ε_{mf} approach. Although beyond the scope of the current study, the modified ε_{mf} approach can be employed to determine the fractions of IPFs that are overcome and remain (represented by $\varepsilon_{mf}/\varepsilon_0$ and $d_{agg,mf}/d_p$, respectively) in the bed. This cannot be accomplished by the generalized U_{mf} deviation and DHR equations and will be addressed in a forthcoming contribution by the authors.

5.3.4 Modified ε_{mf} approach

5.3.4.1 Background and concept

Under individual particle fluidization conditions, the bed expands from ε_0 to ε_{mf} to increase the average mutual particle-particle distance, weaken IPFs, and, subsequently, allow the particles to move and be fluidized [7]. The magnitude of IPFs under these circumstances can be estimated by

the DHR only, e.g., by employing the non-generalized DHR equation, Eq. 5.1. In agglomerating fluidization, however, two elements of IPFs are distinguishable:

- (i) **IPFs acting between adjacent agglomerates or inter-agglomerate forces (yellow arrows in Figure 5.2a):** Considering the agglomerates as fluidizing entities, this element resembles IPFs in individual particle fluidization. Thus, its contribution to the total magnitude of IPFs is signified by $\varepsilon_{mf}/\varepsilon_0$ in Eq. 5.12. These forces must vanish during bed fluidization to allow the movement of agglomerates. This element typically has a subordinate contribution to the total magnitude of IPFs in agglomerating fluidization.
- (ii) **IPFs between primary particles within agglomerates (red arrows in Figure 5.2a and Figure 5.2b):** The term $d_{agg,mf}/d_p$ reflects the contribution of this element to the total magnitude of IPFs in Eq. 5.12. These forces are not overcome during bed fluidization and persist in acting on particles after fluidization. This happens since the average mutual particle-particle distances within the agglomerates remain virtually unchanged during fluidization (Figure 5.2a and Figure 5.2b). This element becomes the major contributor to the total magnitude of IPFs when $IPFs/W_p \gg 1$, i.e., for powders with a typical Geldart group C behaviour for which $d_{agg,mf}/d_p \gg 1$.

Quantification of the magnitude of IPFs by the modified ε_{mf} approach is based on considering a hypothetical minimum fluidization state, where all particles are individually fluidized. This hypothetical state is similar to the individual particle fluidization conditions, where both elements of IPFs are overcome. We call the bed voidage at this state the modified minimum fluidization voidage $\varepsilon_{mf,mod}$. Therefore, the magnitude of IPFs can be quantified by the non-generalized DHR equation if ε_{mf} is replaced by $\varepsilon_{mf,mod}$:

$$\frac{IPFs}{W_p} = \left(\frac{\varepsilon_{mf,mod}}{\varepsilon_0} \right)^3 \frac{1 - \varepsilon_0}{1 - \varepsilon_{mf,mod}} - 1 \quad 5.15$$

Since inter-agglomerate forces vanish at U_{mf} [25], the average distance between fluidizing agglomerates was considered as the required distance to overcome IPFs. This implies that the primary particles would fluidize individually, i.e., the hypothetical minimum fluidization state, if

they were separated by such distance. Accordingly, $\varepsilon_{mf,mod}$ is estimated as if the average mutual particle-particle distance between primary particles was equal to that between fluidizing agglomerates (Figure 5.2c). In the next section, a model will be presented for the estimation of $\varepsilon_{mf,mod}$ based on this strategy.

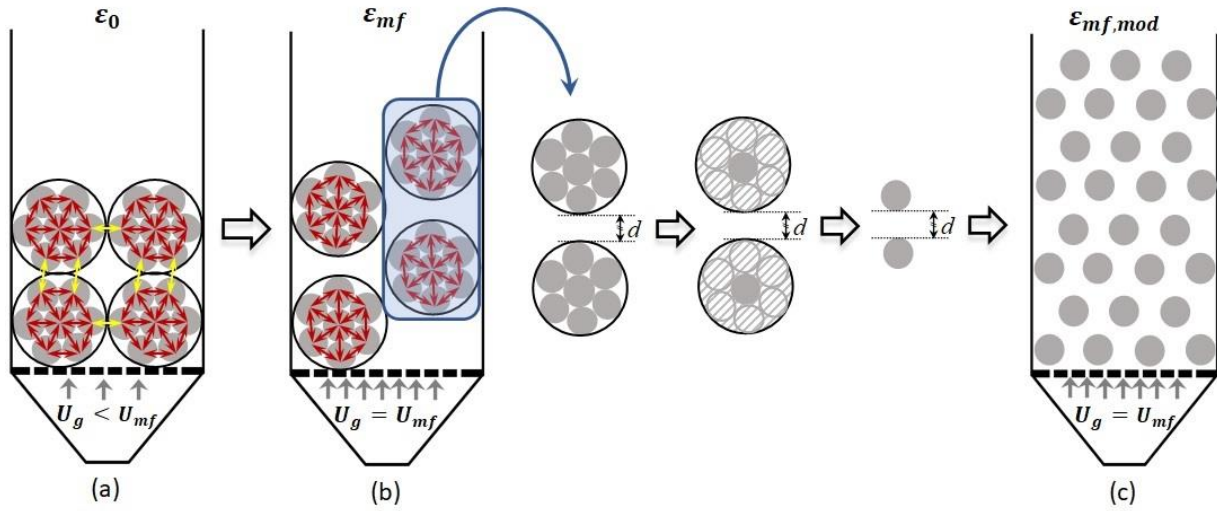


Figure 5.2. Schematic representation of the modified ε_{mf} approach: a) agglomerates at the fixed bed state, b) agglomerates at U_{mf} , and c) hypothetical individual particle fluidization state at U_{mf} .

5.3.4.2 Estimation of $\varepsilon_{mf,mod}$

The following assumptions are adopted to estimate $\varepsilon_{mf,mod}$:

- (i) Identical spherical primary particles with a diameter of d_p form the particulate bed.
- (ii) Agglomerates at minimum fluidization conditions are identical spheres with a diameter of $d_{agg,mf}$.

- (iii) The voidage of an agglomerate at minimum fluidization conditions $\varepsilon_{agg,mf}$ was assumed to be as follows:

$$\varepsilon_{agg,mf} = \varepsilon_{agg,\infty,mf} \left[1 - \left(\frac{d_p}{d_{agg,mf}} \right)^3 \right] \quad 5.16$$

where $\varepsilon_{agg,\infty,mf}$ is the voidage of an extremely large agglomerate at U_{mf} .

- (iv) $\varepsilon_{mf,mod}$ is calculated as if the particles are individually fluidized, leading to a situation where the average mutual particle-particle distance is equal to the average mutual agglomerate-agglomerate distance at U_{mf} . As shown in Figure 5.2, this hypothetical state is achieved by substituting a single primary particle for each agglomerate in the system.

Assumption (iii) is a linear interpolation of $\varepsilon_{agg,mf}$ with the particle to agglomerate volume ratio ($= \left(\frac{d_p}{d_{agg,mf}} \right)^3$) corresponding to the following conditions:

$$\varepsilon_{agg,mf} \rightarrow 0 \text{ if } \frac{d_p}{d_{agg,mf}} \rightarrow 1 \quad 5.17$$

$$\varepsilon_{agg,mf} \rightarrow \varepsilon_{agg,\infty,mf} \text{ if } \frac{d_p}{d_{agg,mf}} \rightarrow 0 \quad 5.18$$

It will be shown in Section 5.5.3 that the voidage of non-fluidizing large agglomerates is approximately equal to the tapped bulk voidage ε_{tap} . $\varepsilon_{agg,\infty,mf}$ is expected to be slightly higher than ε_{tap} owing to the dynamic breakage and formation of agglomerates at U_{mf} .

Experimental minimum fluidization voidage is defined as the ratio of the total gas volume $V_{g,t}$ to the total bed volume at U_{mf} :

$$\varepsilon_{mf} = \frac{V_{g,t}}{V_{g,t} + V_p N_p} \quad 5.19$$

where V_p and N_p are the volume of a particle and the total number of particles in the bed. Therefore:

$$V_{g,t} = \frac{\varepsilon_{mf}}{1 - \varepsilon_{mf}} V_p N_p \quad 5.20$$

$\varepsilon_{mf,mod}$ is defined as:

$$\varepsilon_{mf,mod} = \frac{V_g}{V_g + V_s} \quad 5.21$$

where V_g and V_s are the total gas and solid volumes in the hypothetical individual particle fluidization state. They can be expressed as the following based on assumption (iv) and Figure 5.2:

$$V_g = V_{g,t} - N_{agg,mf} V_{agg,mf} \varepsilon_{agg,mf} \quad 5.22$$

$$V_s = N_{agg,mf} V_p \quad 5.23$$

where $V_{agg,mf}$ and $N_{agg,mf}$ are the volume of an agglomerate and the total number of agglomerates in the bed at minimum fluidization conditions. Considering assumptions (i) and (ii), $V_{agg,mf}$ can be expressed as:

$$V_{agg,mf} = V_p \left(\frac{d_{agg,mf}}{d_p} \right)^3 \quad 5.24$$

$N_{agg,mf}$ can be calculated by dividing the total solids volume by the volume of solids in an agglomerate:

$$N_{agg,mf} = \frac{V_p N_p}{V_{agg,mf} (1 - \varepsilon_{agg,mf})} = \frac{N_p}{(1 - \varepsilon_{agg,mf})} \left(\frac{d_p}{d_{agg,mf}} \right)^3 \quad 5.25$$

By substituting Eqs. 5.20, 5.24, and 5.25 in Eqs. 5.22 and 5.23, we obtain:

$$V_g = \frac{\varepsilon_{mf}}{1 - \varepsilon_{mf}} V_p N_p - \frac{\varepsilon_{agg,mf}}{(1 - \varepsilon_{agg,mf})} N_p V_p \quad 5.26$$

$$V_s = \frac{N_p V_p}{(1 - \varepsilon_{agg,mf})} \left(\frac{d_p}{d_{agg,mf}} \right)^3 \quad 5.27$$

The $\varepsilon_{mf,mod}$ can subsequently be calculated by substituting Eqs. 5.26 and 5.27 in Eq. 5.21:

$$\varepsilon_{mf,mod} = \frac{(\varepsilon_{mf} - \varepsilon_{agg,mf})}{(\varepsilon_{mf} - \varepsilon_{agg,mf}) + (1 - \varepsilon_{mf}) \left(\frac{d_p}{d_{agg,mf}} \right)^3} \quad 5.28$$

Following the above procedure based on assumption (iv), the number of particles in the hypothetical state is equal to the number of agglomerates (not the number of primary particles) in the real fluidization state. However, as expected and expressed in Eq. 5.28, $\varepsilon_{mf,mod}$ is independent of the number of particles or agglomerates. This agrees with the experimental data showing that the bed voidage is practically independent of its height/ inventory [7, 23].

In accordance with expectation, Eq. 5.28 indicates that $\varepsilon_{mf,mod}$ is equal to the experimental ε_{mf} in the case of individual particle fluidization. One will be able to quantify the magnitude of IPFs in agglomerating fluidization with Eq. 5.15 upon the estimation of $\varepsilon_{mf,mod}$. The results of the modified ε_{mf} approach will be employed to cross-check the magnitude of IPFs quantified by the generalized DHR and U_{mf} deviation equations.

5.4 Experimental

Fluidization tests were carried out at ambient conditions in a 5.25 cm diameter transparent Plexiglas fluidized bed with a porous distributor plate. The windbox was filled with glass beads of 5 mm in diameter to help with an even distribution of dried air, as the fluidizing gas, over the unit cross-section. The fluidized bed rig was electrically grounded to avoid accumulation of electrostatic charges, which can change $U_{mf,exp}$ and the fluidization behaviour of powders over time. Three calibrated rotameters were installed on the air inlet line to help accurately adjust the gas flow rate over a wide range of superficial gas velocities, up to 0.16 m/s.

The experiments were carried out with ten solid materials with effective particle diameters ranging from 0.5 to 35 microns. The effective particle diameter, which, for the sake of simplicity, is referred to as the particle diameter in the rest of this document, is defined as the mean particle diameter multiplied by its shape factor [18]. Physical properties of the powders under investigation were reported in Table 2. The relatively narrow size distributions of the powders ensure complying with the assumption of unisize particles adopted for the development of the generalized U_{mf} deviation and DHR equations.

Table 5.2. Physical properties of particles.

Material	Particle density (kg/m ³)	Sieving size (μm)	Sauter mean particle diameter (μm)	Sphericity (-)	Effective particle diameter (μm)	Geldart group behaviour	Acronym
Iron oxide (Fe ₃ O ₄)	4780	<2 ^a	0.6	0.86 ^b	0.5	C	Fe-0.5
Iron oxide (Fe ₂ O ₃)	4895	<5 ^a	1.5	0.69 ^b	1	C	Fe-1
Silicon oxide (SiO ₂)	2634	0.5-10 ^a	4	0.73 ^b	3	C	Si-3
Calcium oxide (CaO)	3161	<15 ^a	8	0.76 ^c	6	C	Ca-6
Lithium iron phosphate (LiFePO ₄)	3585	<38	20	0.71 ^b	14	C	LFP-14
Cornstarch	1502	<38	22	0.81 ^c	18	C	CS-18
Glass beads	2576	<32	20	~1 ^b	20	A	GB-20
Wheat flour	1466	20-45	31	0.85 ^c	26	C	WF-26
Sugar	1591	20-45	36	0.86 ^c	31	C	S-31
Phosphogypsum (CaSO ₄ .2H ₂ O)	1344	32-53	47	0.74 ^c	35	A	PG-35

(a) Data provided by the supplier.

(b) Results from EyeTech particle size and shape analyzer.

(c) Estimated by SEM images.

Densities of particles were measured with a Micromeritics® AccuPyc II 1340 Pycnometer employing helium as the penetrating gas. Laser diffraction particle size analyzers may measure the agglomerate size rather than that of the primary cohesive particles [26]. The wet mode analysis by EyeTech particle size and shape analyzer helps minimize van der Waals and electrostatic forces as it is carried out in the water environment [7]. Consequently, the agglomerates are broken, and the equipment measures the primary particle size. This approach, however, cannot be employed for Ca-6, CS-18, WF-26, S-31, and PG-35, which are soluble in water or form a soft surface and stick together. Hence, SEM images of about 100 primary particles by the HITACHI TM3030Plus scanning electron microscope were analyzed for their size estimation.

Experimental U_{mf} is routinely defined as the superficial gas velocity, where the total bed pressure drop line for the fixed bed state intersects with the virtually constant total bed pressure drop at the fully fluidized state [18]. $U_{mf,exp}$ was calculated from the decreasing velocity pass pressure drop profile according to Richardson's classical method [27]. Total bed pressure drop at each superficial gas velocity U_g was measured by JUMO, 404304/000-414-415-28-298 pressure transducer. The average bed voidage ε_{ave} at each U_g was calculated by:

$$\varepsilon_{ave} = 1 - \frac{m_s/\rho_p}{A L_{ave}} \quad 5.29$$

where ρ_p , m_s , and A are the particle density, bed inventory, and bed cross-sectional area, respectively. L_{ave} is the average bed height and was determined by visual observation of the bed surface. The bed voidage at complete shut down of the gas flow upon its gradual decrease from the fully fluidized state was considered as the loose bulk voidage.

ε_{mf} , as the average bed voidage at minimum fluidization velocity, was calculated upon determining U_{mf} . The tapped bulk voidage was measured by manually tapping the bed for a short time, e.g., 60 seconds, after which the bed voidage remained constant.

The following procedure was applied to estimate the voidage of non-fluidizing large agglomerates $\varepsilon_{agg,\infty}$. A 3 cm X 3 cm metal sheet was covered with a double-sided adhesive tape. Powder

containers were gently tilted such that a few large agglomerates from the surface of the particulate assembly fall on the surface of the adhesive tape from a very close distance. These were nearly spherical agglomerates of Fe-0.5, Fe-1, Si-3, and CaO-6 with diameters ranging from 2 to 7 mm, which were at least three orders of magnitude greater than the corresponding primary particle size. The mass of the agglomerates $m_{agg,\infty}$ were calculated as the difference between the mass of the metal sheet before and after the addition of agglomerates. The average diameters of the agglomerates were measured by SEM images, which helped calculate their volumes $V_{agg,\infty}$. Ultimately, $\varepsilon_{agg,\infty}$ was calculated as:

$$\varepsilon_{agg,\infty} = 1 - \frac{m_{agg,\infty}/\rho_p}{V_{agg,\infty}} \quad 5.30$$

The experimental measurement of $U_{mf,exp}$ together with the bed height, pressure drop, and voidage at this gas velocity, enabled us to estimate the effective agglomerate diameters under minimum fluidization conditions by rearranging the viscous term of the Ergun equation:

$$d_{agg,mf} = \sqrt{\frac{C_{Erg} U_{mf,exp} \mu_g L_{mf} (1 - \varepsilon_{mf})^2}{\varepsilon_{mf}^3 \Delta P_{mf}}} \quad (Re_{p,mf} < 20) \quad 5.31$$

where L_{mf} and ΔP_{mf} are the bed height and pressure drop at $U_{mf,exp}$, respectively.

Agreement between the estimations of Eq. 5.31 and the experimentally measured agglomerate sizes with a high-speed camera was reported in literature [28]. In this work, they were verified through comparison with the agglomerate diameters estimated by an optical fiber probe (see Supplementary Material).

5.5 Results and discussion

In a recent study by the group [7], the relationships between IPFs and ε_0 , ε_{mf} , and $d_{agg,mf}$ for particles with typical behaviour of Geldart group B and coarser Geldart group A were investigated.

The critical observations were presented in Section 5.3.3. This communication is dedicated to the quantification of IPFs for finer particles of Geldart group A and those with Geldart group C behaviour based on their fluidization features. This helps broaden our knowledge about the magnitude of IPFs for particles with a wide range of physical properties and the effects of these forces on the fluidization characteristics of corresponding powders.

5.5.1 Experimental results

The results of the normalized bed pressure drop profiles, which is defined as the total bed pressure drop ΔP divided by the equivalent pressure drop ΔP_{eq} are presented in Figure 5.3.

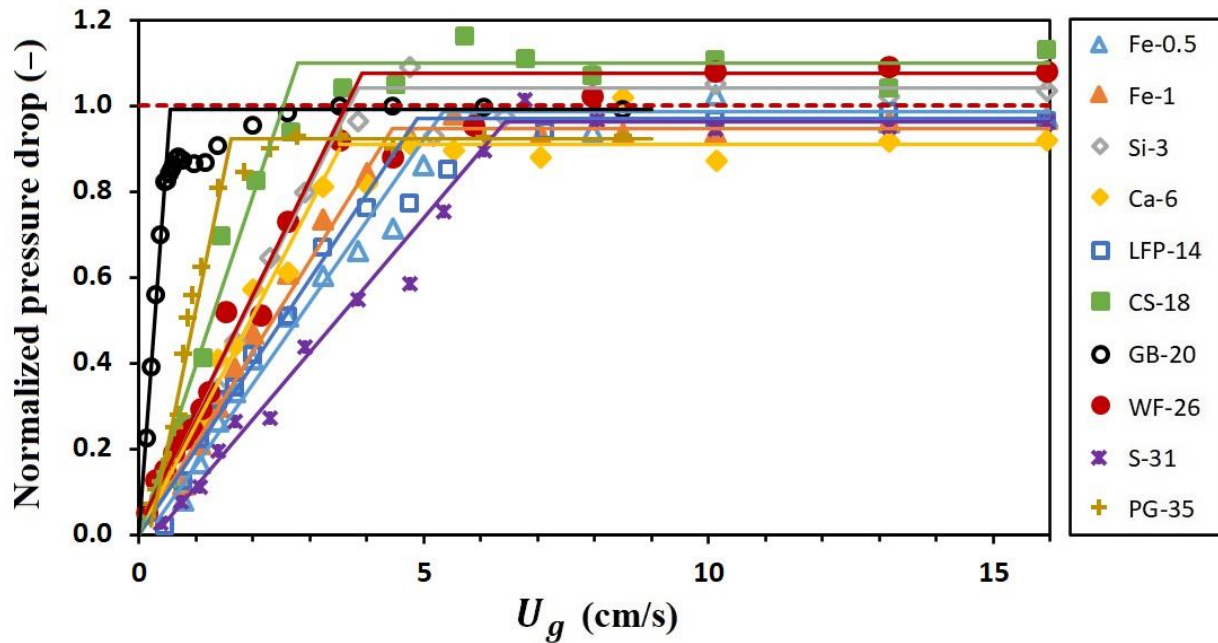


Figure 5.3. Normalized pressure drop profiles in decreasing velocity pass for different particles investigated in this study (lines are representative trendlines for the fixed and fluidized bed states).

The results show that fluidization quality index, i.e., the normalized pressure drop at high gas velocities, varied between 0.91 to 1.10 which was followed by a decrease in the fixed bed state for all powders. The fluidization quality index close to one, similar to the results in Figure 5.3, is regarded as an indication of a proper fluidization behaviour [29]. Slightly greater values for Si-3, CS-18, and WF-26 can be attributed to their higher particle-wall cohesion in comparison with other powders. Formation of a layer of these particles on the inner wall of the bed, which was observed during experiments, can also affect the normalized pressure drop through decreasing the effective bed cross-sectional area.

Selection of pressure drop data points for the trendlines of the fixed and fluidized bed states is a significant uncertainty of the Richardson's method [27] for the estimation of $U_{mf,exp}$. In this study, the values of $U_{mf,exp}$ were calculated as the average of minimum fluidization velocities obtained by different reasonable sets of data points for the fixed and fluidized states. The representative trendlines for the fixed and fluidized beds are illustrated in Figure 5.3. The difference between the maximum and minimum calculated values was considered as the uncertainty in the measurement of $U_{mf,exp}$. The calculated values of $U_{mf,exp}$ were in the range of 0.55 cm/s (for GB-20) to 6.44 cm/s (for S-31). Subsequently, the corresponding normalized pressure drops at $U_{mf,exp}$ were calculated within 0.8 to 0.96 which considered as the values of $F_{fp,mf}$.

The proposed equations remain applicable as long as the adopted assumptions for their developments remain valid and if the degree of uncertainty is acceptable for the user. The former, i.e., the validity of the adopted assumptions, is not always the case for powders with Geldart group C behaviour, in particular, when severe channeling/slugging in the bed results in very erratic pressure drop profiles with no distinct $U_{mf,exp}$. Accordingly, powders with very erratic pressure drop profiles, fluidization quality indices smaller than 0.9, $F_{fp,mf}$ smaller than 0.8 or uncertainties greater than 20% in the measurement of $U_{mf,exp}$ were excluded from our analyses. They include 6-micron FePO_4 , 8-micron color pigment, 6-micron LiFePO_4 , 3-micron Li_2CO_3 , 10-micron Ca(OH)_2 , 15-micron LiPO_3 , 10-micron wheat flour, 12-micron sugar, and 2-micron SiC particles. All powders with Geldart group C behaviour in Figure 5.3 showed channeling at minimum fluidization. The channeling was more pronounced for Si-3 and Ca-6, where channels continued

to appear at higher gas velocities (3 to 5 times $U_{mf,exp}$). For other powders, channels formed at minimum fluidization conditions and transformed into bubbles at higher gas velocities. This behaviour, also known as quasi-fluidization conditions, is reported when $IPFs/W_p$ is in the order of 10-1000 [23], similar to the conditions of most Geldart group C powders in this study (see Figure 5.6). Fe-0.5 and Fe-1 showed a minor degree of channeling. Considering their mean diameters, these powders seem to behave at the boundary of agglomerating particulate fluidization (APF) and agglomerating bubbling fluidization (ABF). APF and ABF are agglomerating fluidization behaviours analogous to Geldart groups A and B behaviours - rather than group C - and are commonly experienced in the fluidization of ultrafine and nanoparticles [8, 9].

Experimental values of ε_0 and ε_{mf} for particles under investigation in this study are shown in Figure 5.4. As expected, it illustrates that the values of ε_{mf} were greater than the corresponding ε_0 for particles with typical Geldart group A behaviour, i.e., GB-20 and PG-35. However, ε_0 and ε_{mf} were very close for the other particles behaving like Geldart group C powders. The similarity of ε_0 and ε_{mf} for these particles is due to the dominance of IPFs over HDFs, which is reflected in the formation of channels [2, 6]. The uncertainties in ε_{mf} were calculated based on the correspondence between ε_{mf} and $U_{mf,exp}$ and considering the uncertainties in the measurement of $U_{mf,exp}$.

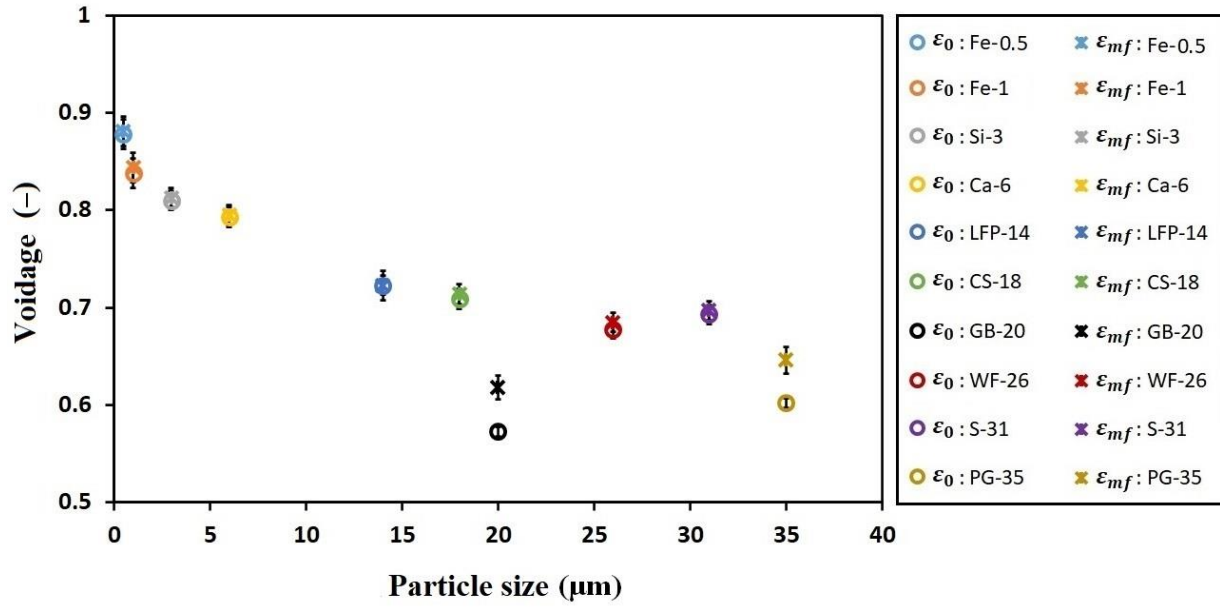


Figure 5.4. Variations of ε_0 and ε_{mf} with d_p for particles under investigation in this study.

The effective agglomerate diameters at $U_{mf,exp}$ are presented in terms of $d_{agg,mf}/d_p$ in Figure 5.5. The uncertainties in the estimation of $d_{agg,mf}/d_p$ and $IPFs/W_p$ (see Figure 5.6) were quantified by applying the error propagation method on Eqs. 5.31 and 5.9. In agreement with reported data in the literature for particles showing Geldart group C behaviour [30], the obtained results show that $d_{agg,mf}$ can be appreciably greater than d_p (one–two orders of magnitude). The ratio of $d_{agg,mf}/d_p$ remained smaller than about 3 for particles showing Geldart group A behaviour (GB-20 and PG-35). The latter observation was previously confirmed for 83-micron FCC particles at high temperatures, where these particles showed fluidization characteristics of Geldart group A [7].

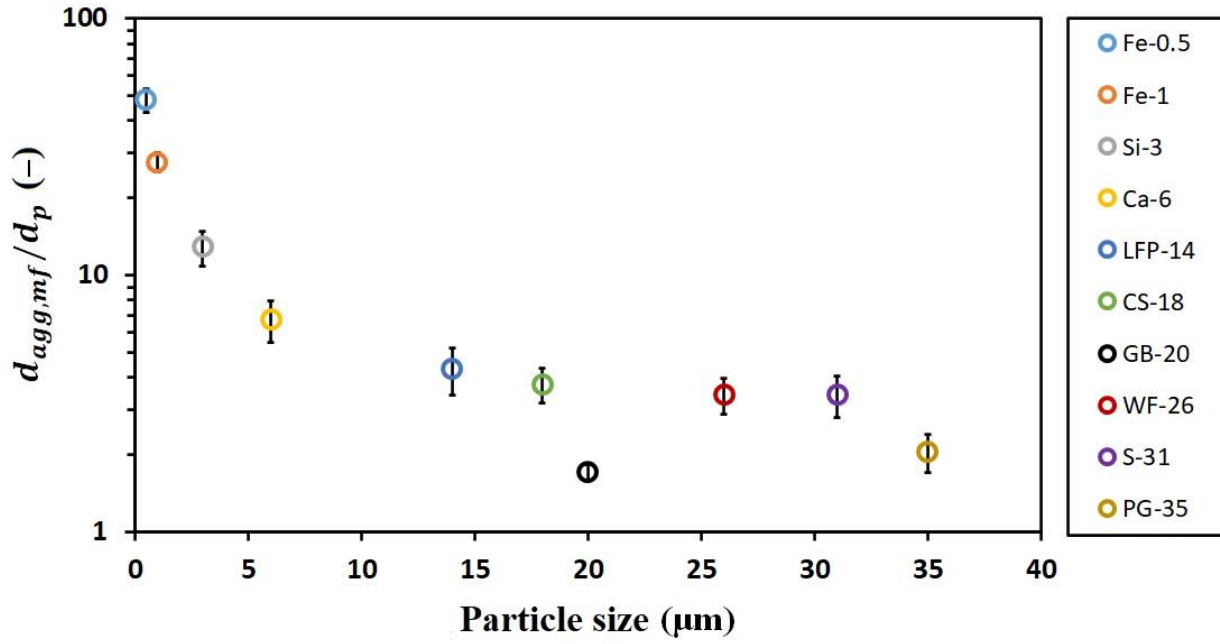


Figure 5.5. Variation of $d_{agg,mf}/d_p$ with d_p for particles under investigation.

The general trends in the variation of ε_0 , ε_{mf} , and $d_{agg,mf}/d_p$ with particle size will be discussed in the following section considering the magnitudes of IPFs.

5.5.2 Quantified magnitudes of IPFs

The $IPFs/W_p$ quantified by the generalized equations are presented in Figure 5.6. Eqs. 5.9 and 5.12 produced the same results since the values of $d_{agg,mf}$ were estimated by Eq. 5.31. Their results can be different if one measures $d_{agg,mf}$ by other techniques. For example, $d_{agg,mf}$ measured by an optical fiber probe (see Supplementary Material) were, on average, 17% different from the estimation of Eq. 5.31. Employing these values in Eq. 5.12 leads to quantifying the magnitude of IPFs about 36% greater than the results of Eq. 5.9. The application of Eq. 5.31 for the estimation of $d_{agg,mf}$ is more relevant in this study as it estimates the mean diameter of

agglomerates throughout the bed rather than their local size, which are measured by the optical fiber probe.

Figure 5.6 indicates that the magnitude of IPFs generally decreased with d_p . It also implies that the magnitudes of IPFs for finer powders, particularly those smaller than 20 μm , were primarily a function of particle size rather than their physical properties. For particles with Geldart group A behaviour, $IPFs$ and W_p were in the same order of magnitude ($IPFs/W_p = 3.1$ and 4.8 for GB-20 and PG-35, respectively). However, for particles showing Geldart group C behaviour, the magnitudes of IPFs were at least an order of magnitude greater than W_p . It will be shown in Sections 5.5.3 and 5.5.5 that the proposed approaches cannot adequately estimate the magnitude of IPFs for ultrafine/nanoparticles (Fe-0.5 and Fe-1 in this study) and must be applied with great caution when the bed voidage is greater than 0.8 (Fe-0.5, Fe-1, and Si-3 in this study). Nonetheless, as shown in Figure 5.6, the magnitude of IPFs for powders with Geldart group C behaviour can be two to three orders of magnitude greater than the weight of these particles. The ranges of quantified magnitudes of IPFs for particles with different Geldart group behaviours in this study are in agreement with corresponding results reported in literature [1, 5, 23]. The observations for ε_0 and ε_{mf} (Figure 5.4), $d_{agg,mf}/d_p$ (Figure 5.5), and corresponding magnitudes of IPFs (Figure 5.6) support our earlier discussions about the relationship between fluidization behaviour and the magnitude of IPFs for Geldart groups A and C powders in Figure 5.1.

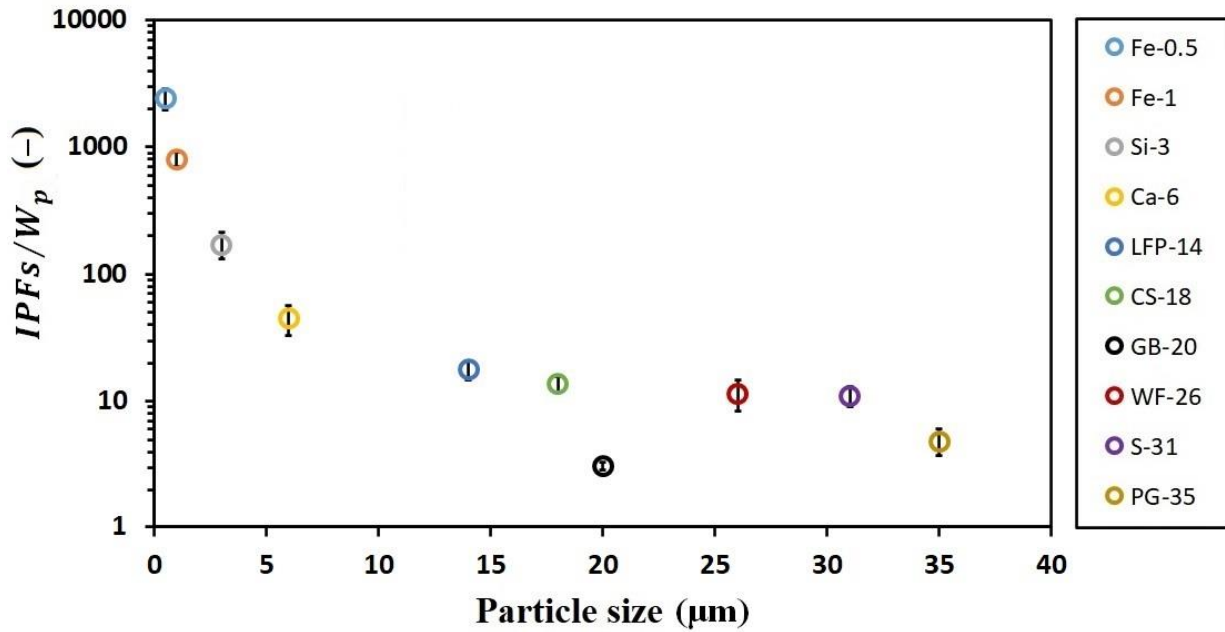


Figure 5.6. Variation of $IPFs/W_p$ quantified by Eq. 5.9 or 5.12 with d_p for particles under investigation.

Since the formation of agglomerates is directly governed by the magnitude of IPFs, similar qualitative trends for $IPFs/W_p$ and $d_{agg,mf}/d_p$ against d_p (Figure 5.5 and Figure 5.6) are expected. The similarity of the observed variations of ε_0 and ε_{mf} with d_p (Figure 5.4) to that of $d_{agg,mf}/d_p$ and $IPFs/W_p$ implies that the bed voidage (ε_0 and ε_{mf}) for the investigated solid materials was primarily impacted by the same factor, i.e., IPFs. A comparison of Figure 5.4 and Figure 5.6 indicates that higher values of ε_0 and/or ε_{mf} correspond to greater magnitudes of IPFs. Considering different physical properties of the explored solid materials, this suggests that the variations of ε_0 and ε_{mf} for fine powders are dominated by their significant magnitudes of IPFs rather than their physical properties. For intermediate-sized and coarse particles with Geldart groups B and D behaviours, where IPFs are insignificant compared to HDFs, the voidage is generally lower in beds of smaller particles [2, 31]. A further decrease in the average particle size to where the bed behaves like Geldart group C leads to an increase in ε_0 and ε_{mf} owing to a

substantial increase in the ratio of IPFs/HDFs [2, 7]. The trend inversion happens when the two effects are comparable, i.e., for particles with Geldart group A behaviour [2, 31].

5.5.3 Verification and validation

This section is dedicated to the verification and validation of the generalized U_{mf} deviation and DHR approaches, i.e., Eqs. 5.9 and 5.12, for the adequate estimation of the magnitude of IPFs in a gas-solid fluidized bed. The performances of the generalized DHR and U_{mf} deviation equations were verified by comparing their results with the criteria proposed in the literature for the magnitude of IPFs in beds with different Geldart group behaviours. The independency of these approaches to the bed dimensions and the gas type (when there is a minimal gas adsorption on the particles' surface) were also categorized as verification proof. The qualitative validation of the generalized approaches was accomplished by the comparison of their results with the quantified IPFs by the modified ε_{mf} approach as well as the van der Waals forces estimated by conventional models.

The order of magnitude of quantified IPFs by Eqs. 5.9 and/or 5.12: Studies have shown that different Geldart group behaviours correspond to specific ranges of relative magnitude of IPFs to HDFs and/or W_p [1, 5, 6]. Particles are expected to behave like Geldart group A when IPFs are comparable with W_p [1, 5], i.e., if $\sim 0.1 < IPFs/W_p < \sim 10$. Fluidization behaviour of Geldart group C powders is governed by IPF [1, 8, 9], which implies that $IPFs/W_p > \sim 10$ for these powders. Quantified IPFs by the generalized U_{mf} deviation and DHR equations for the explored solid materials are in agreement with these criteria.

Effect of bed dimensions on the quantification of IPFs by Eqs. 5.9 and/or 5.12: The magnitude of IPFs as an intrinsic physical property of the particles is independent of the bed dimensions [7]. Thus, the proposed equations for quantifying these forces must lead to similar results when the particles are fluidized in beds with different diameters and/or heights. The parameters in Eqs. 5.9 and 5.12 that need to be measured include $U_{mf,exp}$, ε_0 , and ε_{mf} , d_p , and $d_{agg,mf}$. The bed dimensions do not appear in most correlations for the prediction of these parameters as they are

considered intrinsic characteristics of particles and essentially constant in beds with different dimensions [7, 32]. This implies that the proposed equations are basically independent of the bed diameter and/or height.

The wall effect is partially responsible for the appearance of the overshoot pressure drop at minimum fluidization in small beds [33, 34]. Nevertheless, only a fraction of the overshoot caused by IPFs is reflected in the DHR equation, i.e., the pressure drop corresponding to the difference between ε_0 and ε_{mf} [7]. The generalized DHR equation remains independent of the bed dimension even in small beds since the ratios of $\varepsilon_{mf}/\varepsilon_0$ and $d_{agg,mf}/d_p$ are minimally impacted by the wall effect. However, the wall effect in small beds may result in an additional increase in $U_{mf,exp}$ due to the friction and cohesion between particles and the bed wall. Therefore, it is recommended that $U_{mf,exp}$ is measured in beds with insignificant wall effect for an adequate estimation of the magnitude of IPFs by the generalized U_{mf} deviation equation.

Effect of gas type on the quantification of IPFs by Eqs. 5.9 and/or 5.12: The fluidizing gas can affect the magnitude of IPFs via adsorption on the particles' surface [35-37]. The level of IPFs is minimally impacted by the fluidizing gases if their adsorption on the particles' surface is negligible [7]. Consequently, the proposed equations must yield similar values of IPFs when the bed is fluidized with different gases under the above circumstances. The condition is usually met at ambient pressure and/or high temperature, similar to the fluidization conditions of FCC particles with air and Argon in a recent study by the authors (at atmospheric pressure and temperatures ranging from ambient to 600°C) [7]. The magnitudes of IPFs for these particles at different temperatures calculated by the generalized equations with the experimental data of fluidization with air and Argon are reported in Figure 5.7. In accordance with our expectations, the results indicate that Eqs. 5.9 and/or 5.12 led to similar results (within 14% experimental error) when applied to fluidization with different gases.

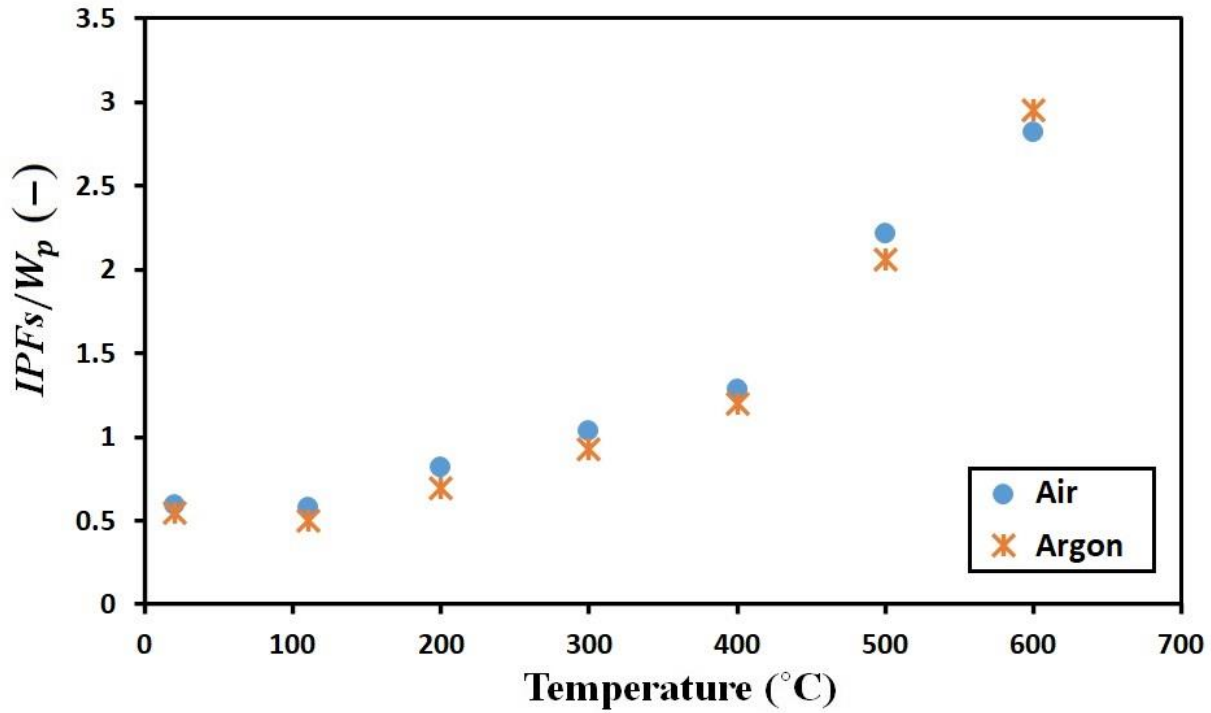


Figure 5.7. Performance of Eqs. 5.9 and/or 5.12 when the bed is fluidized with different gases at atmospheric pressure and/or high temperature. (Experimental data for 83-microns FCC particles was adopted from [7])

Comparison of quantified IPFs by Eqs. 5.9 and/or 5.12 with van der Waals forces: van der Waals forces are the main type of IPFs for fine powders under ambient operating conditions and in a dry environment [38]. Therefore, the obtained IPFs by the generalized approaches in this study must be similar to the prediction of models proposed in the literature for the estimation of the van der Waals forces. However, these models are highly dependent on the particle's surface properties, whose measurements are hardly reliable for fine ($<20\ \mu\text{m}$) and irregular-shaped powders [39], such as particulate samples adopted in this study. Nevertheless, reported data for similar powders (29 microns glass beads) in the study by LaMarche et al. [40] enabled us to estimate the magnitude of van der Waals forces for GB-20. Although this calculation was associated with a high degree of uncertainty, the van der Waals forces calculated by the Hamaker [41] and Rumpf [42] models ($IPFs/W_p = 7.4$ and 11.5 , respectively) are comparable with the results of the generalized DHR

and U_{mf} deviation approaches ($IPFs/W_p = 3.1$), especially considering that the Hamaker and Rumpf models estimate maximum value of van der Waals forces when the separation distance is equated to intermolecular distance ($\approx 4 \text{ nm}$) [43].

Another conclusion can be reached by comparing the IPFs acting on different size glass beads with Geldart group A behaviour (GB-20 in this study, GB-35 for which $IPFs/W_p = 0.84$ at $\varepsilon_0 = 0.53$ (the results are not shown in this communication), and GB-67 in [7]). The conventional models adequately estimate van der Waals forces acting on these particles if the particle's roughness is substituted for its diameter [43, 44]. Therefore, assuming similar surface roughness, pairwise IPFs acting on GB-20, GB-35, and GB-67 (not $IPFs/W_p$) must be comparable. The experimental results are compatible with these theoretical predictions (IPFs = 0.10, 0.15, and 0.16 nN for GB-20, GB-35, and GB-67, respectively). This is a critical finding and helps estimate the magnitude of van der Waals forces for different particle sizes of a given material knowing its value for one. It can also be regarded as a further support for the validity of the proposed approaches.

Comparison of quantified IPFs by Eqs. 5.9 and/or 5.12 with the results of the modified ε_{mf} approach: To estimate the magnitude of IPFs by the modified ε_{mf} approach, one needs to know $\varepsilon_{agg,\infty,mf}$. The values of $\varepsilon_{agg,\infty}$ i.e., the voidages of non-fluidizing large agglomerates were measured for Fe-0.5, Fe-1, Si-3, and Ca-6. The results were close to the corresponding tapped bulk voidages ($\varepsilon_{agg,\infty} = \varepsilon_{tap}^{+0.01}_{-0.04}$). As alluded to earlier $\varepsilon_{agg,\infty,mf}$ is expected to be close to, but slightly greater than, $\varepsilon_{agg,\infty}$ ($\approx \varepsilon_{tap}$) due to the aeration of the bed and the dynamic process of formation and breakage of the agglomerates at U_{mf} [6]. On the other hand, $\varepsilon_{agg,\infty,mf}$ must be smaller than the average bed voidage since the void space between agglomerates (Figure 5.2a and Figure 5.2b) together with that within the agglomerates contribute to the average bed voidage. Therefore:

$$\varepsilon_{tap} \approx \varepsilon_{agg,\infty} < \varepsilon_{agg,\infty,mf} < \varepsilon_0 \approx \varepsilon_{mf} \quad 5.32$$

A comparison between the results of the generalized equations and those of the modified ε_{mf} approach when $\varepsilon_{agg,\infty,mf}$ was equated to ε_{tap} is presented in Figure 5.8. Apart from Fe-0.5, Fe-1,

and Si-3, which will be discussed later, the modified ε_{mf} approach estimated comparable but slightly greater magnitudes of IPFs (with an average of 30 % and a maximum of 44 %) than the results of Eqs. 5.9 and/or 5.12. This is in accordance with our expectation as equating $\varepsilon_{agg,\infty,mf}$ with its lower limit, i.e., ε_{tap} , causes an overestimation of the magnitude of IPFs by the modified ε_{mf} approach.

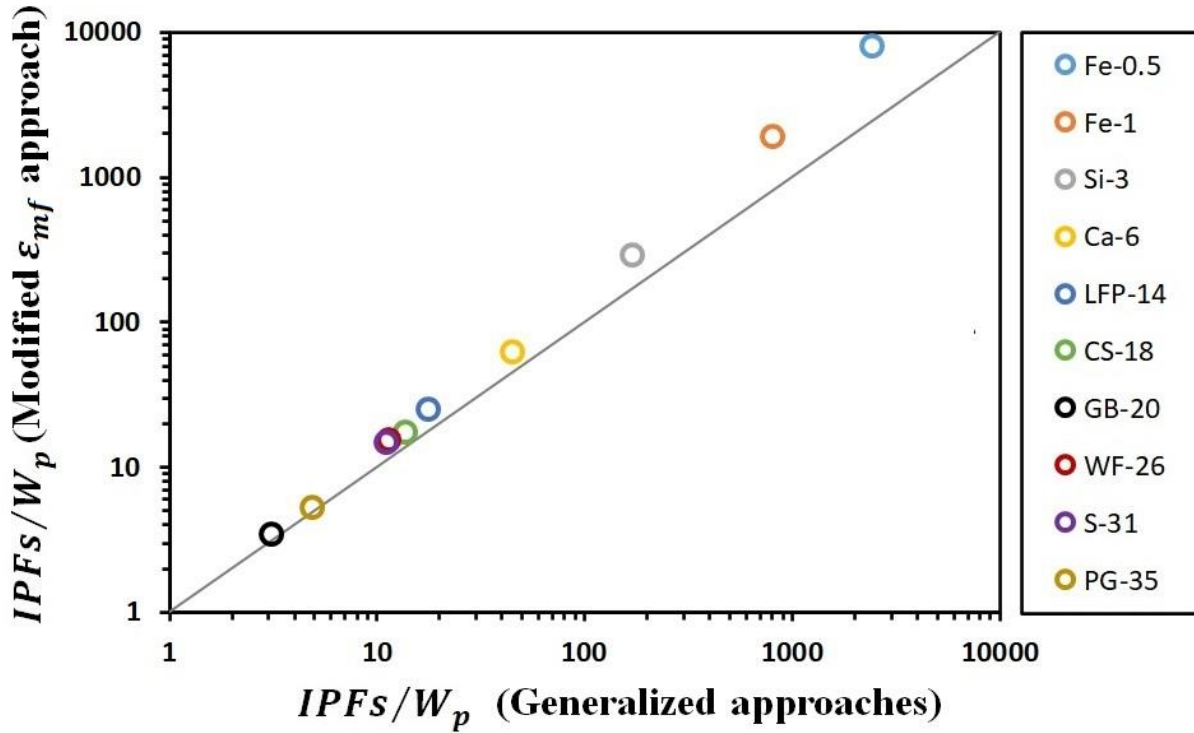


Figure 5.8. Comparison of the results of the modified ε_{mf} and generalized approaches when $\varepsilon_{agg,\infty,mf}$ was equated to ε_{tap} .

The results presented in Figure 5.8 and the above discussion recommend that a more appropriate value of $\varepsilon_{agg,\infty,mf}$ should be employed in the modified ε_{mf} approach for an adequate estimation of the magnitude of IPFs. This value must be between ε_{tap} and ε_0 based on Eq. 5.32. The results suggest that employing the following expression for $\varepsilon_{agg,\infty,mf}$ minimizes the discrepancy between

the results of the modified ε_{mf} approach and the generalized equations for particles other than Fe-0.5, Fe-1, and Si-3 (an average of 5% and a maximum of 9% difference):

$$\varepsilon_{agg,\infty,mf} = 0.75\varepsilon_{tap} + 0.25\varepsilon_0 \quad 5.33$$

As will be discussed in Section 5.5.5, Fe-0.5, Fe-1, and Si-3 fall into the application limits of the proposed approaches, i.e., ultrafine/nanoparticles and/or when the bed voidage is greater than 0.8. Thus, the estimates of the modified ε_{mf} approach were significantly greater than those of the generalized equations in these cases (239%, 139%, and 74% for Fe-0.5, Fe-1, and Si-3, respectively). The discrepancy between the results of the proposed approaches for these particles remains significant (160%, 86%, and 32% Fe-0.5, Fe-1, and Si-3, respectively) even after employing Eq. 5.33 to estimate $\varepsilon_{agg,\infty,mf}$.

5.5.4 Application of the proposed approaches to quantify the magnitude of IPFs at elevated temperatures

The experimental data showing the variations of the DHR and $d_{agg,mf}$ with temperature for 83-microns FCC particles are presented in Figure 5.9a and Figure 5.9b (data adopted from [7]). Results showed that the $d_{agg,mf}$ significantly grows with temperature ($d_{agg,mf}/d_p$ increased from 1.06 to 1.50), while the DHR remained essentially constant (within 1.07–1.13) over the explored range of operating temperature. Comparisons of $U_{mf,exp}$ and $U_{mf,pred}$ estimated at the corresponding ε_0 are presented in Figure 5.9c. As discussed in Section 5.3.3, the deviation of $U_{mf,exp}$ from $U_{mf,pred}|_{\varepsilon_0}$ could be attributed to two factors. The first factor, i.e., the difference between ε_{mf} and ε_0 was partially responsible for this deviation at all temperatures. The second, i.e., the formation of agglomerates became dominant at elevated temperatures as larger agglomerates formed under those conditions. Accordingly, the non-generalized DHR equation underestimates the magnitude of IPFs (Figure 5.9d) since it cannot account for the contribution of $d_{agg,mf}/d_p$ in the total magnitude of IPFs. The underestimation is particularly significant at

elevated temperatures, where $d_{agg,mf}/d_p$ is greater. Results in Figure 5.9d also indicate that the quantified magnitudes of IPFs by the generalized equations agree with those by the modified ε_{mf} approach, which could be considered as further support for the qualitative validation of the proposed approaches.

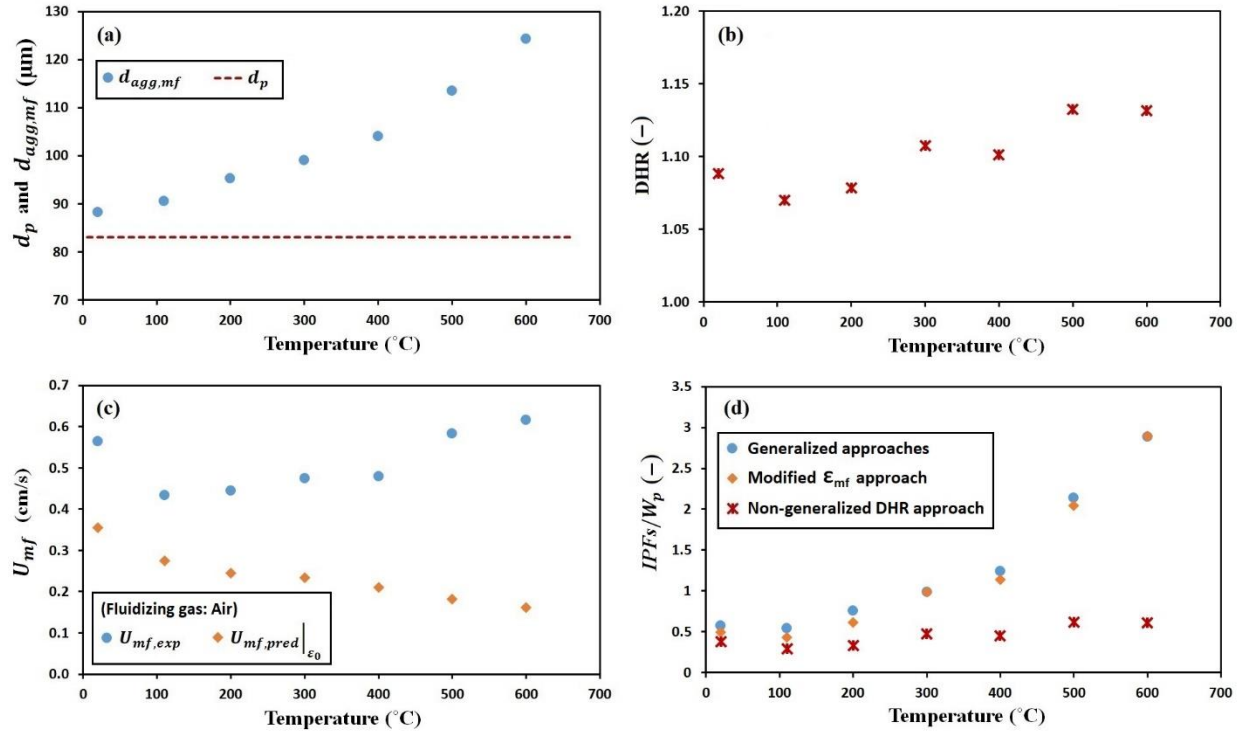


Figure 5.9. Performance of the generalized equations, modified ε_{mf} approach, and non-generalized DHR approach at high temperature. (Experimental data for 83-microns FCC particles was adopted from [7])

5.5.5 Uncertainties and limitations of the proposed approaches

The reliability and accuracy of the proposed approaches in this study for quantifying IPFs in gas-solid fluidized beds depend on the consistency of the assumptions and conditions under which they are developed and employed. These parameters are as follows:

Flow regime: The proposed approaches were developed based on the assumption of a viscous flow regime at U_{mf} ($Re_{p,mf} < 20$), where the inertia term in the Ergun equation can be safely neglected. If this condition is not met, for example, for coarse particles with Geldart group D behaviour, the proposed approaches cannot be employed. According to this background, we did not include Geldart group D behaviour in our discussion in Section 5.3.3, although these particles experience individual particle fluidization and have similar ε_0 and ε_{mf} , similar to Geldart group B behaviour.

Measurements of U_{mf} and ε_{mf} : The general uncertainties associated with the measurement of U_{mf} and ε_{mf} include the uneven distribution of fluidizing gas across the bed cross-section and inaccurate measurement of hydrodynamic parameters such as superficial gas velocity, pressure drop, and bed height, caused by miscalibration or improper placement/installation of the measurement devices, e.g., flowmeters, and pressure transducers. The methods used for the estimation of U_{mf} and ε_{mf} from experimental data can also introduce additional uncertainties. For example, one of the significant uncertainties of the Richardson's method [27] is the selection of the data points for the pressure drop line in the fixed bed state and those for the relatively flat line in the fluidized state. This method is, however, the most widely accepted approach for the measurement of $U_{mf,exp}$ generating reproducible results for particles with different fluidization behaviours and, hence, was employed in this study.

Uncertainties in the measurement of U_{mf} and ε_{mf} for cohesive powders are amplified by their fluidization behaviour. Their erratic and non-reproducible pressure drop profiles lead to imprecise U_{mf} estimations [45]. In addition, channeling causes greater gas flow in the channels compared to non-fluidized regions of the bed. Hence, the actual gas velocity in fluidized regions is greater than the average superficial gas velocity, i.e., the reported value for U_{mf} . Similarly, the bed voidage in the channel region may be greater than the average bed voidage, which is considered as ε_{mf} . Moreover, a layer of powders on the inner wall of the bed can form due to the cohesive forces between fine particles and the bed wall. Not only does this phenomenon make it challenging to obtain an accurate reading of the bed height by visual observation, but it also decreases the effective bed diameter and the weight of the fluidizing material. This, in turn, leads to a systematic

error in the estimation of U_{mf} . The above uncertainties in the measurement of $U_{mf,exp}$ and ε_{mf} for cohesive particles lead to an underestimation of the magnitude of IPFs by the proposed approaches for these powders.

Severe slugging/channeling: In extreme cases with Geldart group C behaviour, the bed pressure drop profile becomes very erratic because of severe slugging or channeling. Consequently, no obvious minimum fluidization condition can be determined. In addition, the bed pressure drop remains appreciably smaller than the equivalent pressure drop leading to a fluidization quality index smaller than about 0.8. The Ergun equation and, hence, the proposed equations become invalid under these circumstances.

Voidages greater than 0.8: The experimental value of C_{Erg} is about 150 at voidages of about 0.4, while its theoretical value is equal to 72 when the bed voidage approaches 1 [19]. C_{Erg} significantly decreases when the bed voidage is greater than 0.8 [19]. Therefore, the Ergun and Carman-Kozeny equations must be cautiously applied for bed voidages greater than 0.8.

The smaller values of C_{Erg} when the bed voidage is greater than 0.8 can partially explain the discrepancy between the quantified IPFs by the proposed approaches for Si-3, Fe-1, and Fe-0.5 particles in this study (see Figure 5.8). The disagreement between the results decreases by 81%, 47%, and 39% for Si-3, Fe-1, and Fe-0.5 particles, respectively, if, rather than a fixed value of 150 for C_{Erg} , we linearly vary it with the bed voidage ($C_{Erg} = -130\varepsilon_{mf} + 202$ corresponding to $C_{Erg} = 150$ at $\varepsilon_{mf} = 0.4$ and $C_{Erg} = 72$ at $\varepsilon_{mf} = 1$). For Si-3 particles, applying this correction to C_{Erg} leads to a 6% difference between the results of the generalized approaches ($IPFs/W_p = 108.2$) and that of the modified ε_{mf} approach when $\varepsilon_{agg,\infty,mf}$ is estimated by Eq. 5.33 ($IPFs/W_p = 114.7$). This recommends that, in this case, employing an appropriate value for C_{Erg} is enough for an adequate estimation of the magnitude of IPFs by the proposed approaches. However, significant discrepancies between the quantified IPFs by the generalized and modified ε_{mf} approaches for Fe-1 and Fe-0.5 remain after the correction of C_{Erg} (46% and 98%, respectively). This suggests that another factor affects the calculation for nanoparticles. This factor will be discussed next.

Nano-powders: The growing inconsistency between the results of the proposed approaches when the particle size decreases to the nanoscale (see Figure 5.8) indicates that these approaches cannot be relied on for a proper estimation of the magnitude of IPFs for ultrafine and nanoparticles.

When large agglomerates - in comparison with the primary particle size - form, the fluidizing gas is expected to principally flow through the void space between agglomerates rather than the space within agglomerates [30]. The effect is known as flow screening and implies that the effective bed voidages at the fixed bed state and U_{mf} are smaller than ε_0 and ε_{mf} , respectively. Hence, for a more realistic estimation of the magnitude of IPFs for ultrafine and nanoparticles, $U_{mf,pred}$ in Eq. 5.9 should be estimated at this effective ε_0 instead of its experimental value. Likewise, employing the effective ε_{mf} in Eq. 5.31 results in the estimation of larger $d_{agg,mf}$, which, in turn, affects the estimation of IPFs by the modified ε_{mf} approach. The quantification of IPFs for nanoparticles remains beyond the scope of this study, while the above discussion provides a framework for future research in the field.

5.6 Conclusion

Two equivalent equations were proposed in this study for the quantification of the magnitude of IPFs in a gas-solid fluidized bed as generalized forms of the U_{mf} deviation and DHR approaches. The generalized U_{mf} deviation equation relates the magnitude of IPFs to the ratio of experimental and theoretical minimum fluidization velocities. The generalized DHR equation correlates the magnitude of IPFs with the agglomerate and particle size ratio in addition to the DHR, i.e., $\varepsilon_{mf}/\varepsilon_0$. The latter equation reduces to its non-generalized form if particles fluidize individually. Similar values of IPFs as the results of the generalized equations were obtained with the non-generalized DHR equation when ε_{mf} was replaced by the voidage of a hypothetical state of the bed, where particles fluidize individually. The bed voidage at this state, $\varepsilon_{mf,mod}$, was estimated as if the average mutual particle-particle distance was similar to the average distance between fluidizing agglomerates.

The results showed that the ratio of $IPFs/W_p$ generally grow with decreasing particle size. While IPFs were in the same order of magnitude as W_p for particles with typical Geldart group A behaviour, they can be three orders of magnitude greater than W_p for fine Geldart group C powders.

The proposed approaches are independent of the source and type of IPFs. They showed promising performances in quantifying IPFs in both individual particle and agglomerating fluidization, either at ambient conditions or elevated temperatures. They can be employed for the development of hydrodynamic correlations, in CFD-DEM simulation, and for the extension of the Geldart classification considering the effect of IPFs. Nevertheless, a limitation analysis of the proposed approaches recommended that they cannot properly estimate the magnitude of IPFs for nanoparticles. Moreover, an appropriate value of the Ergun equation constant must be employed when the bed voidage is greater than 0.8.

Acknowledgements

The authors gratefully acknowledge the financial support provided for this research project by OCP Group and Mitacs through the Mitacs Accelerate program.

5.7 Supplementary Material

5.7.1 On the prediction of minimum fluidization velocity by the Ergun equation

5.7.1.1 Ergun equation

The Ergun [20] equation is widely employed to estimate the pressure drop in packed beds and has been extensively validated by experimental data [19]. Its general form can be expressed as:

$$\Delta P = C_{Erg} \frac{\mu_g}{d_p^2} \frac{L_0(1 - \varepsilon_0)^2}{\varepsilon_0^3} U_g + 1.75 \frac{\rho_g}{d_p} \frac{L_0(1 - \varepsilon_0)}{\varepsilon_0^3} U_g^2 \quad 5.34$$

where C_{Erg} is a constant whose experimental value is about 150 for normal beds having voidages smaller than 0.8. This value sharply decreases at voidages greater than 0.8 and approaches its theoretical value, i.e., 72, when the bed voidage is close to unity [19]. d_p is the particle's mean diameter, which must be replaced by the effective mean diameter for non-spherical particles to account for their shape factor. L_0 and ε_0 are the fixed bed height and its voidage. μ_g , ρ_g , and U_g are the gas viscosity, density, and superficial velocity, respectively.

The first and second terms on the right-hand side of the Ergun equation are viscous and inertia terms, respectively, referring to their relative importance in different flow regimes. The ratio of these two terms can be formulated as:

$$\frac{\text{Inertia term}}{\text{Viscous term}} = \frac{Re_p}{85.7(1 - \varepsilon_0)} \quad 5.35$$

where Re_p is the particle's Reynolds number. Eq. 5.35 indicates that the viscous effect dominates over inertia when Re_p is small. Typically, the inertia term is considered to be negligible when $Re_p < 20$ [18]. This is usually the case for intermediate-sized and fine particles ($d_p < 500 \mu\text{m}$) at superficial gas velocities close to their minimum fluidization velocities. In this case, the Ergun equation can be written as:

$$\Delta P = C_{Erg} \frac{\mu_g}{d_p^2} \frac{L_0(1 - \varepsilon_0)^2}{\varepsilon_0^3} U_g \quad \text{if } Re_p < 20 \quad 5.36$$

5.7.1.2 Minimum fluidization velocity

The experimental minimum fluidization velocity is usually determined with the help of the total bed pressure drop profile in the decreasing velocity pass according to Richardson's classical method [27]. Based on this method, U_{mf} is defined and calculated as the superficial gas velocity, where the bed pressure drop line in the fixed bed state intersects with the virtually constant pressure drop in the fully fluidized state (Figure 5.10). The total bed pressure drop at the fully fluidized

state is nearly equal to the equivalent pressure drop, i.e., the weight of the bed material divided by the bed cross-sectional area [18].

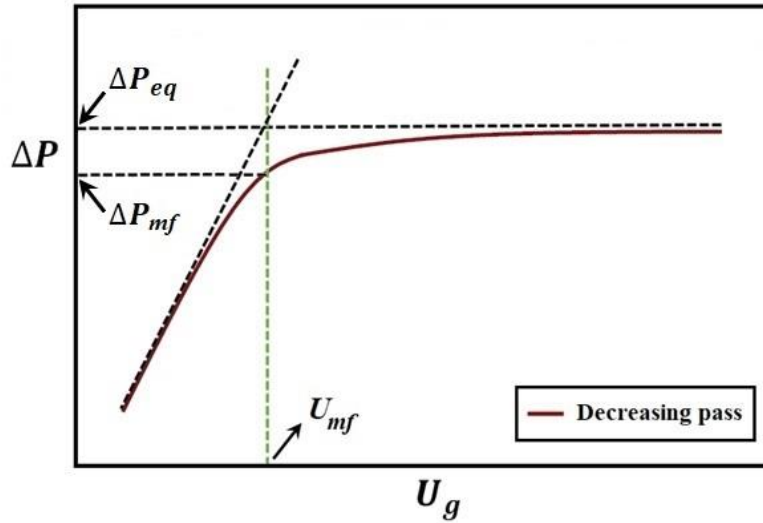


Figure 5.10. Total bed pressure drop profile and estimation of the minimum fluidization velocity by Richardson's classical method [27].

5.7.1.3 Estimation of U_{mf} by Ergun equation

Considering the buoyancy force, the equivalent total bed pressure drop ΔP_{eq} can be written as:

$$\Delta P_{eq} = \frac{(\rho_p - \rho_g)V_{s,tot}g}{A} \quad 5.37$$

where ρ_p and $V_{s,tot}$ are the particle density and total volume of bed solid material, A is the bed cross-sectional area, and g is the gravitational acceleration. $V_{s,tot}$ can be expressed as:

$$V_{s,tot} = AL_0(1 - \varepsilon_0) \quad 5.38$$

Therefore:

$$\Delta P_{eq} = (\rho_p - \rho_g)(1 - \varepsilon_0)L_0g \quad 5.39$$

The bed pressure drop at minimum fluidization ΔP_{mf} is normally smaller than ΔP_{eq} (Figure 5.10) owing to, for instance, the presence of dead/non-fluidized zones in the bed, an appreciable level of cohesive interparticle forces and/or wall friction force in the bed, and improper distribution of fluidizing gas across the bed cross-section. The ratio of ΔP_{mf} and ΔP_{eq} is introduced as the fraction of fluidized particles at minimum fluidization conditions $F_{fp,mf}$ [7]:

$$F_{fp,mf} = \frac{\Delta P_{mf}}{\Delta P_{eq}} \quad 5.40$$

Hence:

$$\Delta P_{mf} = F_{fp,mf}(\rho_p - \rho_g)(1 - \varepsilon_0)L_0g \quad 5.41$$

5.7.1.4 Estimation of U_{mf} for Geldart group B behaviour:

For particles with typical Geldart group B behaviour, the minimum fluidization voidage ε_{mf} is similar to ε_0 . Hence, Eq. 5.36 can be rewritten to estimate ΔP_{mf} as:

$$\Delta P_{mf} = C_{Erg} \frac{\mu_g}{d_p^2} \frac{L_0(1 - \varepsilon_0)^2}{\varepsilon_0^3} U_{mf} \quad 5.42$$

By equating Eqs. 5.41 and 5.42 and assuming $C_{C-K} = \frac{C_{Erg}}{F_{fp,mf}}$, the Carman-Kozeny [17] equation is derived:

$$U_{mf} = \frac{d_p^2}{C_{C-K}} \frac{(\rho_p - \rho_g)}{\mu_g} \frac{\varepsilon_0^3}{(1 - \varepsilon_0)} g \quad 5.43$$

The value of C_{C-K} is usually approximated at 180, considering that $C_{Erg} = 150$ and $F_{fp,mf}$ is usually about 0.8-0.9.

5.7.1.5 Estimation of U_{mf} in individual fluidization for Geldart group A behaviour:

Geldart group A behaviour is characterized by the appearance of an overshoot in the bed pressure drop profile at U_{mf} . This feature is due to the greater value of ε_{mf} in comparison with the corresponding ε_0 for this class of powders [7, 11]. If particles with Geldart group A behaviour fluidize individually, which is usually the case in the fluidization of coarser particles in this group at ambient conditions [7], Eqs. 5.41 and 5.42 can be rewritten as:

$$\Delta P_{mf} = F_{fp,mf}(\rho_p - \rho_g)(1 - \varepsilon_{mf})L_{mf}g \quad 5.44$$

$$\Delta P_{mf} = C_{Erg} \frac{\mu_g}{d_p^2} \frac{L_{mf}(1 - \varepsilon_{mf})^2}{\varepsilon_{mf}^3} U_{mf} \quad 5.45$$

By equating Eqs. 5.44 and 5.45, the modified version of the Carman-Kozeny equation for the prediction of U_{mf} in the individual particle fluidization of Geldart group A behaviour is derived as:

$$U_{mf} = \frac{d_p^2}{C_{C-K}} \frac{(\rho_p - \rho_g)}{\mu_g} \frac{\varepsilon_{mf}^3}{(1 - \varepsilon_{mf})} g \quad 5.46$$

The only difference between Eq. 5.46 and Eq. 5.43 is that ε_0 in Eq. 5.43 was replaced by ε_{mf} in Eq. 5.46. Formisani et al. [11] highlighted the difference between ε_{mf} and ε_0 , in particular, at elevated temperatures for particles showing Geldart group A behaviour. It has been shown that the ratio of $\varepsilon_{mf}/\varepsilon_0$ for these powders is generally greater at higher temperatures due to the enhanced magnitude of IPFs under those operating conditions [7]. It has been also demonstrated that the Carman-Kozeny equation can adequately predict U_{mf} for these particles when ε_{mf} is substituted for ε_0 [11, 22].

5.7.1.6 Estimation of U_{mf} in agglomerating fluidization for Geldart group A and C behaviour:

If primary particles fluidize in the form of agglomerates with an effective diameter of $d_{agg,mf}$, its value must be substituted for d_p in the Ergun equation for an adequate estimation of the total bed pressure drop. This remark changes Eq. 5.45 to:

$$\Delta P_{mf} = C_{Erg} \frac{\mu_g}{d_{agg,mf}^2} \frac{L_{mf}(1 - \varepsilon_{mf})^2}{\varepsilon_{mf}^3} U_{mf} \quad 5.47$$

However, the alternative expression for ΔP_{mf} i.e., Eq. 5.44 remains unchanged as the weight of the agglomerates is equal to the weight of the primary particles. By equating Eqs. 5.44 and 5.47, we obtain:

$$U_{mf} = \frac{d_{agg,mf}^2 (\rho_p - \rho_g)}{C_{C-K} \mu_g} \frac{\varepsilon_{mf}^3}{(1 - \varepsilon_{mf})} g \quad 5.48$$

Eq. 5.48 is the general form of the Carman-Kozeny equation. It was employed in this study to develop the generalized DHR equation for the estimation of the magnitude of IPFs. It is not, however, a predictive correlation for estimation of the minimum fluidization velocity as ε_{mf} and $d_{agg,mf}$ are not known prior to the fluidization test.

5.7.2 Measurement of agglomerate diameter by an optical fiber probe

A reflective optical fiber probe was used for the estimation of the agglomerate diameter at minimum fluidization conditions. The probe was installed at about 6 cm above the distributor plate along the bed axis, i.e., the fixed bed's center. The fiber probe has a diameter of 4.7 mm with 72 emitting and receiving plastic fiber strands. Details of the probe fabrication and specs are reported elsewhere [12].

Prior to experiments with cohesive powders that form agglomerates, the probe was calibrated in beds of the same material but with slightly larger particles, which fluidize individually. The correlation between the bed voidage ε and the voltage associated with the reflected light V for each particle size is given by Cui et al. [46] as follows:

$$\varepsilon = 1 - \frac{a(1 - \varepsilon_{mf})(V - V_0)}{(1 + a)(V_{mf} - V_0) - (V - V_0)} \quad 5.49$$

where ε_{mf} and V_{mf} are the minimum fluidization voidage and the corresponding voltage, V_0 is the voltage in the empty bed (which was set to zero in our experiments), and a is a constant related to the material properties, usually about 0.4 [46].

The optical fiber probe was calibrated in the individual particle fluidization of glass beads (34, 50, 67, and 92 μm) and silica sand particles (35, 54, 75, and 97 μm). Finally, the agglomerate sizes of GB-20 and Si-3 were estimated by measuring the voltage associated with the reflected light in the corresponding beds and with the help of their calibration data. The results showed that GB-20 and Si-3 form agglomerates of 38 μm and 50 μm , respectively. They agree with the measurement with the help of the Ergun equation (34 μm and 39 μm for GB-20 and Si-3, respectively) with less than 23% difference.

The above agreement between the agglomerate sizes estimated by the optical fiber probe and that with the Ergun equation has the following implications: (i) the Ergun equation can adequately estimate the agglomerate size and (ii) for the Ergun equation and, subsequently, the Carman-Kozeny equation, to remain valid in agglomerated particle fluidization, one must replace the particle diameter with the agglomerate size. Therefore, we employed the Ergun equation to estimate the values of $d_{agg,mf}$ in the main manuscript. We also derived the general form of the Carman-Kozeny equation in Section 5.7.1.6 of this document to be used in the development of the generalized DHR equation.

5.8 Nomenclature

Acronyms

ABF	Agglomerating bubbling fluidization
APF	Agglomerating particulate fluidization
Ca-6	6-micron calcium oxide (CaO) particles
CS-16	16-micron cornstarch particles
CFD-DEM	computational fluid dynamics coupled with discrete element method
DHR	dynamic Hausner ratio
FCC	fluid cracking catalyst
Fe-0.5	0.5-micron iron oxide (Fe_3O_4) particles
Fe-1	1-micron iron oxide (Fe_2O_3) particles
GB-20	20-micron glass beads
GB-35	35-micron glass beads
GB-67	67-micron glass beads
HDFs	hydrodynamic forces
IPFs	interparticle forces
LFP-14	14-micron lithium iron phosphate (LiFePO_4) particles
PG-35	35-micron Phosphogypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) particles
SEM	Scanning Electron Microscope
Si-3	3-micron silicon oxide (SiO_2) particles
S-31	31-micron sugar particles

WF-26 26-micron wheat flour particles

Symbols

A	bed cross-sectional area (m ²)
C_{C-K}	constant in the Carman–Kozeny equation (-)
C_{Erg}	constant in the Ergun equation (-)
$d_{agg,mf}$	effective agglomerate diameter at minimum fluidization conditions (m)
d_p	particle diameter (m)
$F_{fp,mf}$	fraction of fluidized particles at minimum fluidization conditions (-)
g	gravitational acceleration (m/s ²)
$IPFs$	interparticle forces acting on each particle at loose bulk state (N)
L_{ave}	average bed height at a given gas velocity (m)
L_{mf}	bed height at minimum fluidization conditions (m)
L_0	bed height at fixed bed state (m), see Supplementary Material
m_s	bed inventory (kg)
$N_{agg,mf}$	total number of agglomerates in the bed at minimum fluidization (-)
N_p	total number of particles in the bed (-)
Re_p	particle Reynolds number (-), see Supplementary Material
$Re_{p,mf}$	particle Reynolds number at minimum fluidization conditions (-)
U_g	superficial gas velocity (m/s)

U_{mf}	minimum fluidization velocity (m/s)
$U_{mf,exp}$	experimental minimum fluidization velocity (m/s)
$U_{mf,exp,ind}$	experimental minimum fluidization velocity in individual particle fluidization conditions (m/s)
$U_{mf,exp,agg}$	experimental minimum fluidization velocity in agglomerating fluidization conditions (m/s)
$U_{mf,pred}$	minimum fluidization velocity predicted by the Carman-Kozeny equation (m/s)
$U_{mf,pred} _{\varepsilon_0}$	minimum fluidization velocity predicted by the Carman-Kozeny equation employing ε_0 as the bed voidage (m/s)
$U_{mf,pred} _{\varepsilon_{mf}}$	minimum fluidization velocity predicted by the Carman-Kozeny equation employing ε_{mf} as the bed voidage (m/s)
$U_{mf,pred} _{\varepsilon_{mf,amb}}$	minimum fluidization velocity predicted by the Carman-Kozeny equation employing $\varepsilon_{mf,amb}$ as the bed voidage (m/s)
$U_{mf,pred} _{\varepsilon_{mf}&d_{agg,mf}}$	minimum fluidization velocity predicted by the Carman-Kozeny equation employing $\varepsilon_{mf,amb}$ as the bed voidage and $d_{agg,mf}$ as the size of the fluidizing entities (m/s)
$V_{agg,mf}$	volume of an agglomerate at minimum fluidization (m ³)
V_g	total gas volume in the hypothetical individual particle fluidization state (m ³)
$V_{g,t}$	total gas volume in a bed at minimum fluidization (m ³)
V_p	volume of a particle (m ³)
V_s	total solid volume in the hypothetical individual particle fluidization state (m ³)
$V_{s,total}$	total volume of particles in the bed (m ³), see Supplementary Material

W_p weight of a single particle (N)

Greek letters

$\Delta IPFs$ thermally induced IPFs acting on each particle (N)

$\Delta IPFs|_{ind}$ magnitude of thermally induced IPFs under individual particle fluidization conditions (N)

ΔP bed pressure drop (Pa)

ΔP_{eq} equivalent bed pressure drop (Pa)

ΔP_{mf} bed pressure drop at minimum fluidization conditions (Pa)

ε_0 loose bulk voidage (-)

ε_{ave} average bed voidage at a given gas velocity (-)

$\varepsilon_{agg,mf}$ voidage of an agglomerate at minimum fluidization (-)

$\varepsilon_{agg,\infty,mf}$ voidage of an extremely large agglomerate at minimum fluidization (-)

$\varepsilon_{agg,\infty}$ voidage of a non-fluidizing large agglomerate (-)

ε_{mf} minimum fluidization voidage (-)

$\varepsilon_{mf,amb}$ minimum fluidization voidage at ambient conditions (-)

$\varepsilon_{mf,mod}$ modified minimum fluidization voidage (-)

ε_{tap} tapped bulk voidage (-)

μ_g gas viscosity (Pa. s)

ρ_g gas density (kg/m³)

ρ_p particle density (kg/m³)

CHAPTER 6 ARTICLE 3: EXTENSION OF GELDART CLASSIFICATION BASED ON THE MAGNITUDE OF INTERPARTICLE FORCES

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Submitted on 17 July 2023, Chemical Engineering Journal

Highlights:

- Variation of Geldart groups A and C boundary with interparticle forces (IPFs).
- IPFs are an order of magnitude greater than the particle weight at Geldart A-C.
- IPFs are an order of magnitude smaller than the particle weight at Geldart A-B.
- Agglomerates are three times greater than the particle size at Geldart A-C.
- Transition between Geldart groups A and C happens at a voidage of 0.6-0.7.

6.1 Abstract

The boundary between Geldart groups A and C is formulated based on the magnitude of interparticle forces (IPFs). It was postulated that the transition from group A to C happens if the interstitial gas velocity at minimum fluidization exceeds the particle terminal velocity. The

proposed model led to novel equations correlating the magnitude of IPFs and the corresponding agglomerate size at this boundary to the bed voidage at minimum fluidization. The proposed boundary was validated by experimental data from literature, including those under an enhanced level of IPFs. The transition between Geldart groups A and C typically occurs when minimum fluidization voidage is 0.6-0.7, corresponding to IPFs about an order of magnitude greater than the weight of a particle and agglomerates about three times larger than the particle size. A comparison of the weight of particles at the boundary between groups A and C and between groups A and B suggests that IPFs are about an order of magnitude smaller than the weight of a particle for the latter. It was shown that, for a given material, particle diameter at the boundary between Geldart groups A and C is proportional to the cube root of interparticle forces. The variation of the boundary between groups A and C with IPFs were presented in a 3D graph.

Keywords: Gas-solid fluidized bed, Hydrodynamics, Interparticle forces, Geldart classification, A-C Boundary, A-B boundary.

6.2 Introduction

Geldart classification [1] has been widely used to predict powders' fluidization behaviour. Based on the mean particle diameter d_p and the difference between the solid and gas densities, $(\rho_p - \rho_g)$, the Geldart chart categorizes particulate materials into four groups with distinct fluidization characteristics. From the heaviest to the lightest, they include groups D, B, A, and C forming spoutable, bubbling, aeratable, and cohesive beds, respectively.

The Geldart classification is based on experimental data limited to fluidization with air at ambient temperature and atmospheric pressure. Typical modifications for including fluid properties in the Geldart chart are based on Archimedes number, which is defined as the ratio of gravitational and viscous forces, and dimensionless density, i.e., $(\rho_p - \rho_g)/\rho_g$ [2-5]. These modifications have shown limited success in the prediction of boundaries between groups D, B, and A, where hydrodynamic and gravitational forces are dominant [4, 5]. Neither the Geldart classification nor the above-mentioned modifications can take into account the effects of the variation of interparticle

forces (IPFs) on the fluidization behaviour. Hence, they fail to effectively predict the fluidization behaviour of a particulate system under an enhanced or diminished level of IPFs [6, 7]. This is a critical challenge under several circumstances. The most practical example of these situations is the design and operation of beds of fine to moderate-sized powders under elevated operating conditions. While these powders typically show Geldart group A or B behaviour at ambient conditions, IPFs induced by elevated operating conditions might shift their behaviour to group C [6, 8-10]. This phenomenon can lead to undesirable characteristics, such as the underperformance of the bed and shut down of the unit in extreme cases. This common situation in the design and operation of catalytic fluidized bed reactors highlights the necessity of an accurate model for predicting Geldart group behaviours under an enhanced level of IPFs, particularly at the boundary between groups A and C (A-C).

IPFs are in the same order of magnitude as the apparent weight of a particle W_p for particles with Geldart group A behaviour [7, 11-13] and govern the fluidization behaviour of group C powders [11, 13-15]. Hence, the proposed criteria for the transition between these two groups (Table 6.1) are either based directly on the magnitude of IPFs [5, 11, 16] or parameters that represent IPFs [17-20]. These parameters include Hausner ratio (HR) [18], angle of repose (AOR) [19], experimental minimum fluidization velocity $U_{mf,exp}$ [17], and specific surface area index $\hat{S}_e$ [20].

Table 6.1. Proposed criteria in literature for the transition from Geldart group A to C behaviour.

Reference	Criteria
Baerns [16] ^a	$\frac{W_p}{W_p + IPFs} < 5 \times 10^{-2}$ (equivalent to $\frac{IPFs}{W_p} > 19$)
Molerus [11] ^b	$10 \frac{(\rho_p - \rho_g) d_p^3 g}{F_{vdw}} < 10^{-2}$ (equivalent to $\frac{IPFs}{W_p} > \sim 5800$)
Chaouki et al. [17]	$U_g > U_t$ at $F_d = W_p + IPFs$ (or $U_{mf,exp} > U_t$)

Geldart et al. [18]	$HR > 1.25 \text{ to } 1.4$
Zhao et al. [19]	$AOR > 35^\circ \text{ to } 40^\circ$
Wang [20]	$\hat{S}_e = \frac{1}{d_p(\rho_p - \rho_g)} > 25$
Mostoufi [5] °	$\frac{IPFs}{W_p} > \frac{50}{Ar^{0.583}} + 0.71 \quad (\text{equivalent to } \frac{IPFs}{W_p} > \sim 100)$

- The criterion corresponds to the quasi-fluidization conditions [16] (see Section 6.4.1).
- The equivalent $IPFs/W_p$ was estimated considering that the resultant IPFs acting on a particle is equal to the product of F_{vdw} and coordination number (Eq. 6.15). The coordination number was estimated by Eq. 6.16 assuming $\varepsilon_{mf} = 0.65$.
- The equivalent magnitude of IPFs is an average for typical powders close to the Geldart A-C boundary ($1000 < \rho_p < 3000 \text{ kg/m}^3$, $15 < d_p < 30 \mu\text{m}$, fluidization with air at ambient conditions).

To evaluate the magnitude of IPFs, studies listed in Table 6.1 adopted different ex-situ approaches, i.e., models and experimental procedures developed to evaluate IPFs outside a fluidized bed. For example, Baerns [16] approximated the total IPFs acting on particles by the powder's angle of slide, Molerus [11] estimated van der Waals forces by the Krupp model [21], and Mostoufi [5] considered the sum of theoretical estimates of different types of IPFs. Consequently, the proposed magnitudes of IPFs corresponding to the A-C boundary differ by orders of magnitude ($IPFs/W_p = 19$ [16], 5800 [11], and 100 [5]). This extensive range of the proposed $IPFs/W_p$ for the Geldart A-C boundary implies that the adopted approaches for the quantification of IPFs are not applicable to the conditions of the aerated particulate assembly in a gas-solid fluidized bed.

In this study, we employed two recently proposed equations for the in-situ quantification of the magnitude of IPFs in a gas-solid fluidized bed by the group [13] to develop a fundamental model

for the boundary between Geldart groups A and C. The performance of the proposed model was evaluated by the available data in literature, including those under an enhanced level of IPFs.

6.3 Theory

6.3.1 Background and assumptions

We recently proposed generalized " U_{mf} deviation" and "DHR (dynamic Hausner ratio)" equations [13] for quantification of the total magnitude of IPFs in gas-solid fluidized beds. The former employs the concept of a delayed minimum fluidization velocity in the presence of IPFs and is formulated as follows [13]:

$$\frac{IPFs}{W_p} = \frac{U_{mf,exp}}{U_{mf,pred}} - 1 \quad 6.1$$

Eq. 6.1, i.e., the U_{mf} deviation equation correlates the magnitude of IPFs to the ratio of experimental minimum fluidization velocity $U_{mf,exp}$ and the prediction of the Carman-Kozeny [22] equation $U_{mf,pred}$ as its theoretical value in the absence of IPFs. $U_{mf,pred}$ in Eq. 6.1 must be calculated by:

$$U_{mf,pred} = \frac{d_p^2}{C} \frac{\rho_p - \rho_g}{\mu_g} g \left(\frac{\varepsilon_0^3}{1 - \varepsilon_0} \right) \quad 6.2$$

where d_p and ρ_p are particle diameter and density, ρ_g and μ_g are the gas density and viscosity, and ε_0 is the bed voidage in the fixed bed state. g is the gravitational acceleration, and C is the constant of the equation, usually 180 [22].

The deviation of $U_{mf,exp}$ from its theoretical value can be attributed to the following reasons [13]: (i) the greater value of minimum fluidization voidage ε_{mf} from ε_0 and, (ii) the formation of agglomerates. The DHR equation correlates these parameters to the magnitude of IPFs as follows [13]:

$$\frac{IPFs}{W_p} = \left(\frac{d_{agg,mf}}{d_p} \right)^2 \left(\frac{\varepsilon_{mf}}{\varepsilon_0} \right)^3 \left(\frac{1 - \varepsilon_0}{1 - \varepsilon_{mf}} \right) - 1 \quad 6.3$$

where $d_{agg,mf}$ is the effective agglomerate size at minimum fluidization conditions. W_p is the apparent weight of a particle, i.e., the difference between gravitational and buoyant forces. For simplicity, it is called the weight of a particle throughout this document.

The ratio of $\varepsilon_{mf}/\varepsilon_0$ in Eq. 6.2 indicates IPFs that must be overcome to fluidize the bed. The agglomerate size ratio $d_{agg,mf}/d_p$, on the other hand, represents IPFs that remain acting on particles even after fluidization. The former is more important for Geldart A particles, while $d_{agg,mf}/d_p$ becomes dominant in the case of Geldart group C powders, where ε_{mf} and ε_0 become similar due to channeling at U_{mf} [13].

The adopted assumptions in this study to model the boundary between Geldart groups A and C are as follows:

- (i) Particles are identical spheres.
- (ii) The gas flow at minimum fluidization follows the viscous flow regime ($Re_{p,mf} < 20$, where $Re_{p,mf}$ is the particle Reynolds number at minimum fluidization).
- (iii) The ratio of IPFs and W_p is estimated by Eq. 6.1 or Eq. 6.3.
- (iv) ε_{mf} is close to ε_0 for particles with Geldart group C behaviour, including those at A-C.
- (v) Channeling, as the distinct characteristic of Geldart group C behaviour, happens if the interstitial gas velocity at U_{mf} exceeds the particle terminal velocity.

6.3.2 The magnitude of IPFs at A-C

From assumption (v), A-C corresponds to:

$$\frac{U_{mf,exp}}{\varepsilon_{mf}} = U_t \quad 6.4$$

where U_t is the terminal fall velocity of a particle and can be estimated by the following equation under the viscous flow regime [23]:

$$U_t = \frac{d_p^2}{18} \frac{\rho_p - \rho_g}{\mu_g} g \quad 6.5$$

Dividing both sides of Eq. 6.4 by $U_{mf,pred}$ yields:

$$\frac{U_{mf,exp}}{U_{mf,pred}} = \frac{\varepsilon_{mf} U_t}{U_{mf,pred}} \quad 6.6$$

By replacing the left-hand side of Eq. 6.6 with Eq. 6.1, the magnitude of IPFs at A-C is obtained as:

$$\left. \frac{IPFs}{W_p} \right|_{A-C} = \frac{\varepsilon_{mf} U_t}{U_{mf,pred}} - 1 \quad 6.7$$

Substitution of $U_{mf,pred}$ and U_t from Eqs. 6.2 and 6.5 into Eq. 6.7 results in:

$$\left. \frac{IPFs}{W_p} \right|_{A-C} = \frac{10 \varepsilon_{mf} (1 - \varepsilon_0)}{\varepsilon_0^3} - 1 \quad 6.8$$

As $\varepsilon_0 \approx \varepsilon_{mf}$ from assumption (iv), we have:

$$\left. \frac{IPFs}{W_p} \right|_{A-C} = \frac{10 (1 - \varepsilon_{mf})}{\varepsilon_{mf}^2} - 1 \quad 6.9$$

Eq. 6.9 expresses A-C as a relationship between the magnitude of IPFs and ε_{mf} . It suggests that $IPFs/W_p$ can take different values depending on the value of ε_{mf} . This feature of the proposed

model shows that the proposed A-C is material dependent as ε_{mf} is an intrinsic property of the particulate assembly.

The magnitude of IPFs corresponding to A-C, Eq. 6.9, and the corresponding agglomerate size ratio, Eq. 6.11, which will be derived in Section 6.3.3, are plotted in Figure 6.1. It shows that for a powder with a smaller ε_{mf} the transition between Geldart groups A and C happens at a greater magnitude of IPFs. For example, the transition occurs at $IPFs/W_p = 10.1$ when $\varepsilon_{mf} = 0.6$ and at $IPFs/W_p = 5.1$ if $\varepsilon_{mf} = 0.7$.

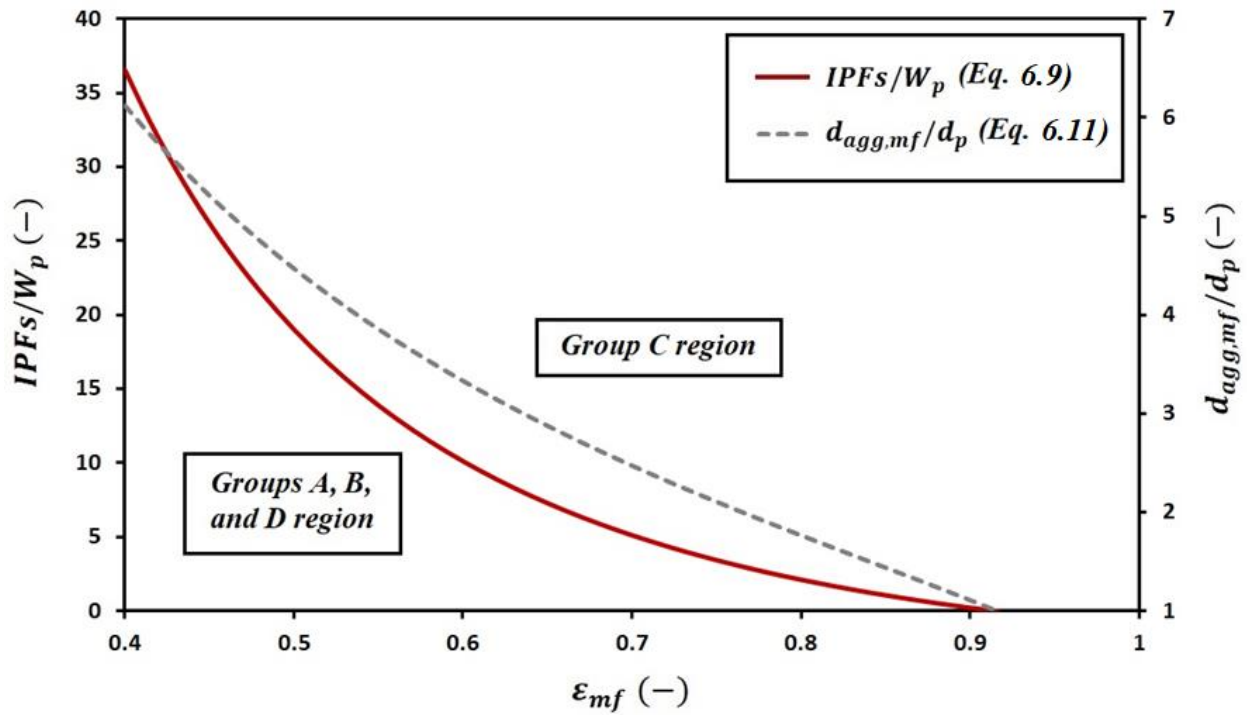


Figure 6.1. Variation of $IPFs/W_p$ with ε_{mf} at A-C boundary and the corresponding agglomerate size ratio.

Figure 6.1 suggests that powders with ε_{mf} greater than about 0.92 will fall into the Geldart group C region irrespective of their magnitudes of IPFs. These high voidages are typical of ultrafine

particles and nanoparticles [14, 15]. In accordance with the prediction of Eq. 6.9, these cohesive solids are commonly classified as Geldart group C. Nanoparticles might, however, show fluidization behaviour of Geldart group A or B, also known as agglomerating particulate fluidization (APF) and agglomerating bubbling fluidization (ABF), respectively [14, 15]. The prediction of ABF and APF fluidization behaviours remains beyond the scope of the current study.

6.3.3 Agglomerate size at A-C

As mentioned in assumption (iv), ε_0 and ε_{mf} are similar for particles with Geldart group C behaviour. Thus, Eq. 6.3 for these powders reduces to:

$$\frac{IPFs}{W_p} = \left(\frac{d_{agg,mf}}{d_p} \right)^2 - 1 \quad 6.10$$

Replacing Eq. 6.10 into Eq. 6.9 results in:

$$\left. \frac{d_{agg,mf}}{d_p} \right|_{A-C} = \sqrt{\frac{10(1 - \varepsilon_{mf})}{\varepsilon_{mf}^2}} \quad 6.11$$

One can employ Eq. 6.11 to estimate the agglomerate size ratio at minimum fluidization conditions at A-C. Figure 6.1 shows that $d_{agg,mf}/d_p$ decreases with ε_{mf} with a qualitatively similar trend as $IPFs/W_p$. This is expected since IPFs are responsible for the formation of agglomerates. It also indicates that $IPFs/W_p$ and $d_{agg,mf}/d_p$ corresponding to A-C depend on the value of ε_{mf} . Experimental results from the literature, however, show that the transition typically happens when ε_{mf} is about 0.6-0.7 (Figure 6.3). For this range of ε_{mf} , $IPFs$ are about an order of magnitude greater than W_p and a $d_{agg,mf}/d_p$ of about 3 is expected. These findings will be discussed in Section 6.4.1.

6.3.4 Variation of A-C with IPFs

W_p can be expressed as:

$$W_p = \frac{\pi}{6} d_p^3 (\rho_p - \rho_g) g \quad 6.12$$

Replacing W_p in Eq. 6.9 and rearranging the equation for d_p at A-C leads to:

$$d_p|_{A-C} = \left[\frac{6 IPFs \varepsilon_{mf}^2}{\pi (\rho_p - \rho_g) g [10 (1 - \varepsilon_{mf}) - \varepsilon_{mf}^2]} \right]^{1/3} \quad 6.13$$

Eq. 6.13 expresses A-C as a relationship between d_p , $\rho_p - \rho_g$, $IPFs$, and ε_{mf} . This equation can be regarded as the generalized form of the Geldart A-C as it accounts for the effect of $IPFs$ and ε_{mf} on A-C in addition to the original Geldart A-C, which is a relationship merely between d_p and $\rho_p - \rho_g$.

$IPFs$ and ε_{mf} are intrinsic properties of the particulate system and are influenced by many factors, including operating conditions and material physical properties, such as the Hamaker constant and surface asperities. Hence, the presence of $IPFs$ and ε_{mf} in Eq. 6.13 explains the large uncertainty in the boundary between Geldart groups A and C. The effect of material properties on the van der Waals forces and, consequently, on $d_p|_{A-C}$ will be further discussed in Section 6.4.2.

The effects of operating conditions on $\rho_p - \rho_g$ and ε_{mf} are limited in comparison with $IPFs$, which can change in orders of magnitude for a given particulate system. Representative examples where the level of IPFs can significantly change include operating at temperatures greater than minimum sintering temperatures of particles, where IPFs substantially grow [24, 25], and water-solid fluidization, where IPFs vanish in the water environment [14, 26]. Therefore, in comparison with IPFs, the variations of other parameters in Eq. 6.13 can be neglected under these circumstances. Accordingly, for a given material, the ratio of $d_p|_{A-C}$ in two conditions and the ratio of $IPFs$ in these conditions can be correlated. Considering ambient as one condition, such analysis of Eq. 6.13 results in:

$$d_p|_{A-C} \approx d_p|_{A-C,ambient} \times \left(\frac{IPFs}{IPFs_{amb}} \right)^{1/3} \quad 6.14$$

Variation of $d_p|_{A-C}$ with $IPFs/IPFs_{amb}$ based on Eq. 6.14 is presented in Figure 6.2. For plotting Figure 6.2, $d_p|_{A-C,ambient}$ was assumed to be equal to the Geldart A-C curve. In other words, it was assumed that the Geldart A-C adequately predicts this boundary at ambient conditions. Hence, the Geldart A-C can be viewed as the intersection of the plane of $IPFs/IPFs_{amb} = 1$ and the surface representing Eq. 6.14 in Figure 6.2.

Figure 6.2 shows that the A-C shifts to the right by enhancing IPFs. For example, while A-C at ambient conditions is experienced for particles with $d_p \approx 20 \mu m$ and $\rho_p - \rho_g > 1000 kg/m^3$, it shifts to $d_p \approx 40 \mu m$ when $IPFs$ increase tenfold. This shift in A-C implies that a $30 \mu m$ particle, which behaves like Geldart group A at ambient conditions, will show group C behaviour under this enhanced IPFs condition. In contrast, the A-C shifts to $d_p \approx 10 \mu m$ if $IPFs$ decrease about tenfold. Both observations are supported by the experimental results in literature [6, 8-10, 26].

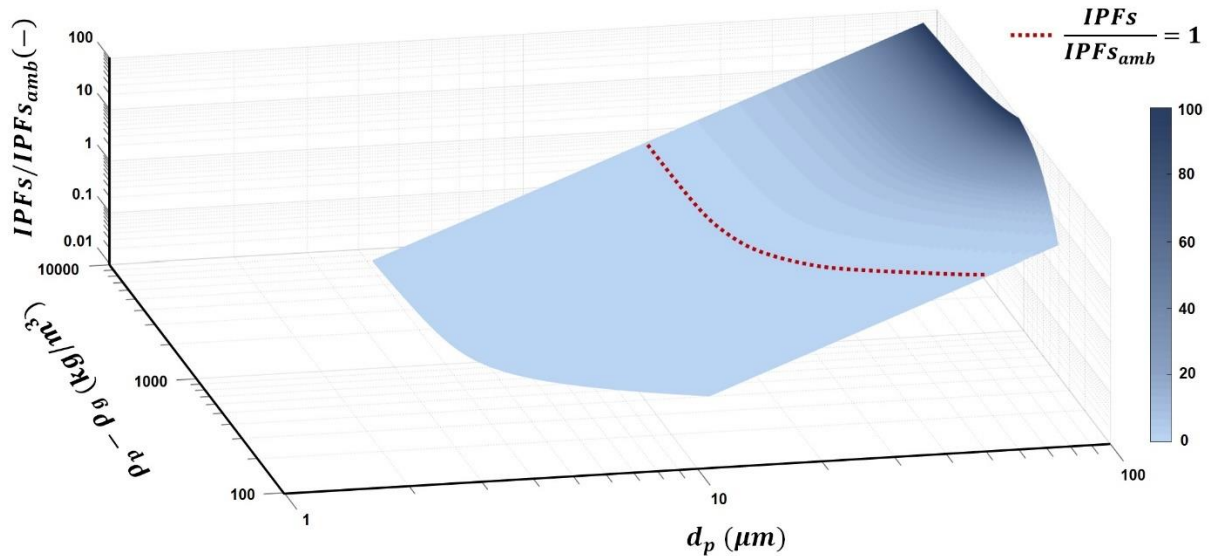


Figure 6.2. Variation of Geldart A-C with respect to IPFs.

By increasing IPFs, a point will be reached where particles with Geldart group B behaviour will also show characteristics of Geldart group C. For instance, particles with $\rho_p - \rho_g > 2000 \text{ kg/m}^3$ and $d_p \approx 90 \text{ }\mu\text{m}$ will behave like group C if $IPFs/IPFs_{amb} \approx 100$.

The van der Waals and electrostatic forces are conversely proportional to the dielectric constant of the medium between particles [7, 14]. Therefore, IPFs in the water environment are about 0.01 times smaller than air. This means that Figure 6.2 may represent the A-C boundary in water-solid fluidization systems when $IPFs/IPFs_{amb} \approx 0.01$. In agreement with experimental data [26], it predicts that the A-C significantly shifts to the left ($d_p \approx 5 \text{ }\mu\text{m}$ for particles with $\rho_p - \rho_g > 1000 \text{ kg/m}^3$). Accordingly, many particles in the original Geldart group C region behave like group A if fluidized with water.

The differences between liquid-solid and gas-solid fluidization are ignored in plotting Figure 6.2 and the corresponding discussion on the variation of the A-C with IPFs. Hence, the proposed water-solid A-C must be regarded as a qualitative analysis of the variation of A-C with IPFs rather than an accurate model. In addition, the uncertainties of the Geldart A-C at ambient conditions for different materials must be considered for a reliable prediction of its variation with IPFs.

6.4 Discussion

6.4.1 Performance of the proposed A-C model

The performance of Eq. 6.9 in the prediction of the boundary between Geldart groups A and C is presented in Figure 6.3. Considering assumption (v) for the development of Eq. 6.9 (see Section 6.3.1), powders with quasi-fluidization behaviour, which show channeling behaviour at U_{mf} but can be fluidized at higher gas velocities [16], are considered as Geldart group C throughout this study.

Data points in Figure 6.3 are experimental data from literature [1, 7, 10, 13, 16, 25, 27-37] for which the magnitudes of IPFs were quantified by U_{mf} deviation or DHR equations. They cover a wide range of conditions, including ambient and elevated temperatures and pressures, fluidization under different humidity levels, sound-assisted fluidization, with uncoated and coated particles and different gases as the fluidizing medium. Detailed information on the data employed in Figure 6.3 is presented in the supplementary material.

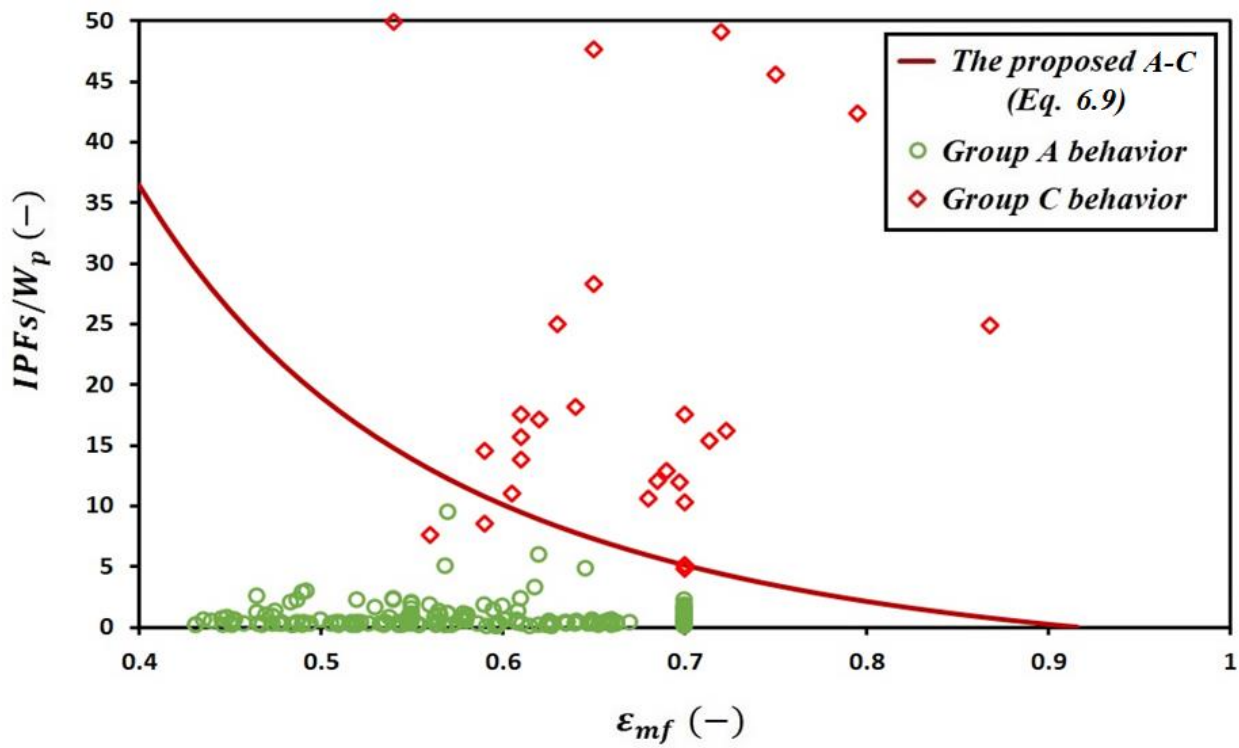


Figure 6.3. Performance of the proposed A-C model based on available data in literature.

Results presented in Figure 6.3 show that the proposed model in this study, i.e., Eq. 6.9, can effectively predict the transition between Geldart groups A and C. No particulate system with $\varepsilon_{mf} > 0.7$ was found in literature to show Geldart group A behaviour, while powders with $\varepsilon_{mf} < 0.6$ rarely showed Geldart group C behaviour. Experimental data in Figure 6.3 reveal that the

transition between Geldart groups A and C typically occurs at $0.6 < \varepsilon_{mf} < 0.7$ corresponding to $IPFs/W_p \approx 5 - 10$ and $d_{agg,mf}/d_p \approx 2.5 - 3.3$. These findings agree with qualitative results in literature showing the dominance of IPFs over gravitational and hydrodynamic forces for Geldart group C powders [38].

The proposed criterion for A-C, i.e., Eq. 6.9, imposes no limitation on the value of ε_{mf} and, subsequently, the magnitude of IPFs. The occurrence of the transition at a certain range of these parameters, however, implies a connection between ε_{mf} and the magnitude of IPFs. Research has shown that the bed voidage, including loose bulk, minimum fluidization, and dense phase voidages, increases with IPFs [7, 10, 28]. By forming larger agglomerates at greater levels of IPFs, the increase in the bed voidage is principally due to the contribution of inter-agglomerate voidage. In extreme cases, e.g., in beds of ultrafine particles and nanopowders, the bed voidage can reach values as high as about 0.99 [15]. This implies that particles experience a significant level of IPFs in beds with high voidage ($\varepsilon_{mf} > 0.8$), where they form large agglomerates. Therefore, these particulate systems must show Geldart group C behaviour as they are located well above the proposed A-C in Figure 6.1. On the other hand, low bed voidage ($\varepsilon_{mf} < 0.5$) generally indicates a small magnitude of IPFs. This, in turn, suggests that particulate systems with low voidage behave like Geldart group B or A as they are located well below the proposed A-C in Figure 6.1. Accordingly, the transition between Geldart groups A and C must happen between these two extremes. At the transition point, ε_{mf} corresponding to Eq. 6.11 matches its dictated value by the contribution of the inter-agglomerate voidage. The presented qualitative discussion can be employed in future research to develop a fundamental model for the variation of the bed voidage with IPFs.

6.4.2 Reconstructing Geldart A-C at ambient conditions

In this section, the proposed A-C is analyzed under conditions where van der Waals forces are the main type of IPFs. These conditions include fluidization at ambient conditions with dry air, i.e.,

the situation under which the Geldart classification was proposed. IPFs under such circumstances can be expressed as:

$$IPFs \approx k F_{vdW} \quad 6.15$$

where k is the coordination number and, under minimum fluidization conditions, can be estimated by [12]:

$$k = 1.61 \varepsilon_{mf}^{-1.48} \quad 6.16$$

F_{vdW} is pairwise van der Waals forces. We previously showed that the Hamaker model provides close values to the quantified IPFs by generalized U_{mf} deviation and DHR equations when particle asperity size, r , is substituted for its radius [7, 13]:

$$F_{vdW} = \frac{r H}{12 a_s^2} \quad 6.17$$

where H is the Hamaker constant, and a_s is the separation distance between neighboring particles. Maximum F_{vdW} is achieved when a_s is in the order of molecular diameter, i.e., about 4×10^{-10} m. By replacing Eqs. 6.15-6.17 in Eq. 6.13, we obtain:

$$d_p|_{A-C} = \left[\frac{0.805 r H \varepsilon_{mf}^{0.52}}{\pi a_s^2 (\rho_s - \rho_g) g [10 (1 - \varepsilon_{mf}) - \varepsilon_{mf}^2]} \right]^{1/3} \quad 6.18$$

Eq. 6.18 is a special case of Eq. 6.13, where van der Waals forces dominate IPFs. Assuming typical values for the parameters in Eq. 6.18, the upper and lower bounds of Geldart A-C at ambient conditions were estimated and presented in Figure 6.4. Unlike Molerus' approach [11], no fitting parameter or empirical constant was employed in Eq. 6.18. The prediction of Eq. 6.18 for the upper and lower bounds is dominated by the range of the Hamaker constant and asperity size as they can largely vary compared to the other parameters in Eq. 6.18. This emphasizes the material dependency of A-C when van der Waals forces dominate IPFs and explains the large uncertainty in the Geldart A-C at ambient conditions.

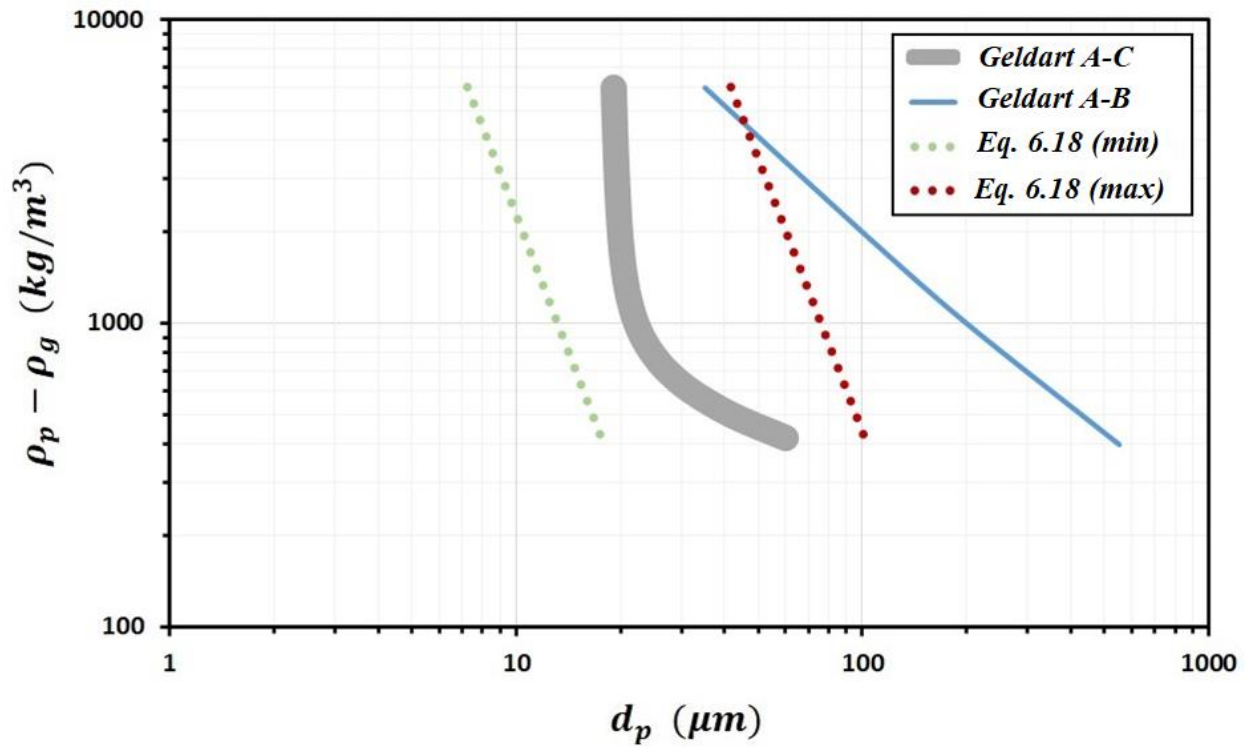


Figure 6.4. Maximum and minimum values of Eq. 6.18 as upper and lower bounds of the Geldart A-C boundary at ambient conditions. ($1 \times 10^{-20} < H < 1 \times 10^{-19}j$, $1 \times 10^{-8} < r < 1 \times 10^{-7}m$, $0.6 < \varepsilon_{mf} < 0.7$, $10^{-10} < a_s < 5 \times 10^{-10}m$).

6.4.3 Boundary between Geldart groups A and B

Although the current study is focused on the boundary between Geldart groups A and C and its relationship with the magnitude of IPFs, a comparison with the boundary between Geldart groups A and B (A-B) might be insightful. While A-C corresponds to the dominance of IPFs, A-B is identified as the point where IPFs are unimportant in comparison with W_p and hydrodynamic forces [11]. Below are two criteria in literature proposed for A-B based on the magnitude of IPFs: Molerus' criterion for A-B can be expressed as [11]:

$$\frac{F_{vdW}}{W_p} = 6.25 \quad 6.19$$

By replacing F_{vdW} from Eq. 6.15 and assuming $\varepsilon_{mf} \approx 0.45$ for the estimation of the coordination number at A-B, Eq. 6.19 can be reformulated as:

$$\frac{IPFs}{W_p} \approx 33 \quad 6.20$$

The proposed criterion for A-B in the study by Mostoufi [5] is as follows:

$$\frac{IPFs}{W_p} = \frac{20}{Ar^{0.662}} + 0.07 \quad 6.21$$

For the fluidization of typical particles at A-B ($1000 < \rho_p < 3000 \text{ kg/m}^3$, $70 < d_p < 210 \mu\text{m}$) with air at ambient conditions, Eq. 6.21 results in $IPFs/W_p \approx 5.3$ on average.

The above two criteria are inconsistent as they suggest different magnitudes of IPFs at A-B. They also violate the initial definition of A-B by Molerus [11] as they show that IPFs are significantly greater than W_p . As alluded to earlier, these inconsistencies originated from the application of improper approaches for the quantification of IPFs in gas-solid fluidized beds.

The findings of the current study on the magnitudes of IPFs at A-C, which are quantified by the DHR and U_{mf} deviation equations, can help estimate the magnitude of IPFs at A-B. W_p of typical particles ($1000 < \rho_p < 3000 \text{ kg/m}^3$) on the boundary between Geldart groups A and B are, on average, more than 100 times greater than those on A-C. Assuming that IPFs are independent of d_p at ambient conditions, $IPFs/W_p \approx 10$ at A-C, and the ratio of W_p at A-B to that at A-C (≈ 100), the magnitude of IPFs at A-B can be estimated as:

$$\left. \frac{IPFs}{W_p} \right|_{A-B} \approx 0.1 \quad 6.22$$

The magnitude of IPFs for particles with Geldart group B behaviour is smaller than 0.1 according to the above analysis and, hence, unimportant in their fluidization behaviour. This conclusion agrees with the observations and explains why the Geldart group B region remains largely beyond the upper bound of Eq. 6.18 at ambient conditions (Figure 6.4).

6.4.4 Variation of A-C boundary with temperature for a given material

As Eq. 6.18 implies, van der Waals forces for micron-sized particles are a function of asperity size rather than particle diameter [24, 39]. Therefore, under certain conditions where van der Waals forces dominate IPFs, Eq. 6.13 can be employed to approximate $d_p|_{A-C}$ for a given material under the same conditions. To this end, IPFs must be known for a particle of the same material under the same conditions. IPFs can be quantified for a particle with an arbitrary diameter, as IPFs are independent of d_p . However, it is preferred to be one with Geldart group A behaviour for which the uncertainties in the measurement of $U_{mf,exp}$ and, consequently, the quantified IPFs are relatively insignificant.

The conditions where the above approach for the estimation of $d_p|_{A-C}$ can be adopted include operating at elevated temperatures up to the minimum sintering temperature of particles. The quantified IPFs for 67-micron glass beads and 83-micron FCC particles at temperatures ranging from ambient to 600°C [7] enabled us to apply this approach to these materials. The estimated $d_p|_{A-C}$ at different temperatures in Figure 6.5 show that $d_p|_{A-C}$ at ambient conditions is close to the Geldart A-C boundary (about 21 and 31 μm for glass beads and FCC, respectively). This can be regarded as a qualitative verification of the adopted approach. By increasing the temperature above 100°C, IPFs grow and shift A-C to the right. At the highest temperatures for each powder material, $d_p|_{A-C}$ reaches about twice its values at ambient conditions.

The knowledge of the transition point between Geldart groups A and C under elevated temperatures is critical for the appropriate design and operation of gas-solid fluidized beds. The

presented approach has the potential to help adequately predict the A-C boundary under these conditions.

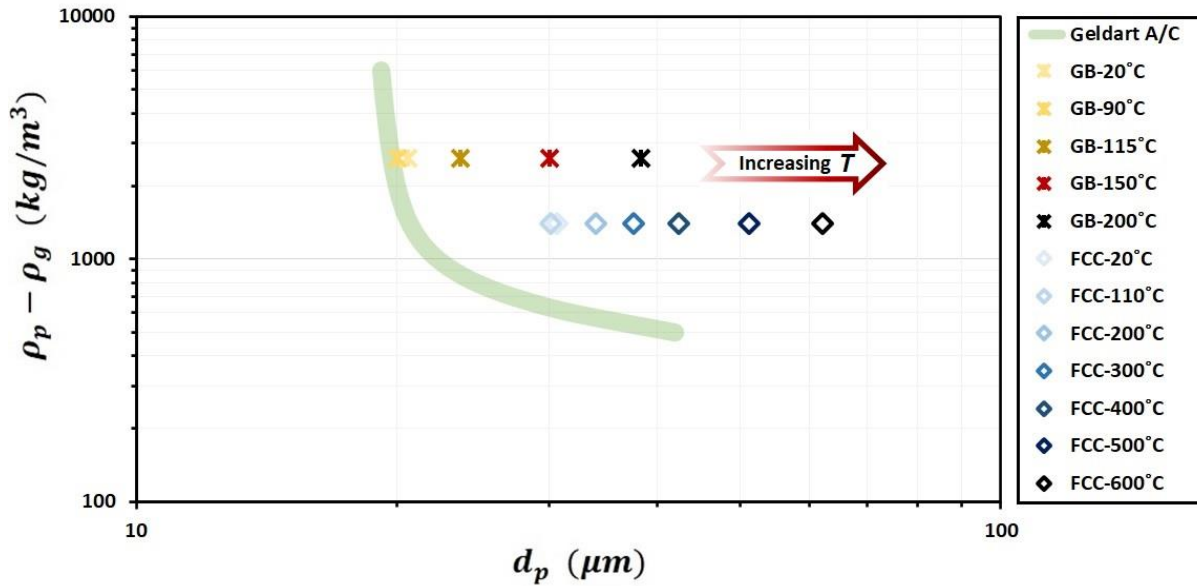


Figure 6.5. Predicted A-C for glass beads and FCC particles at elevated temperatures.

6.4.5 Limitations of the proposed model

Any deviation from the adopted assumptions for developing the proposed A-C model (see Section 6.3.2) imposes a degree of uncertainty on its application. Common scenarios are the wide size distribution of particles and the deviation of the constant of the Carmen-Kozeny equation from 180, for example, when the bed voidage is greater than 0.8 [13, 40]. Extreme cases where the DHR and U_{mf} deviation equations, as the basis of the proposed A-C model, are invalid include the non-viscous flow regime and nanoparticle fluidization, which are discussed below.

Non-viscous flow regime: Both DHR and U_{mf} deviation equations were developed under the viscous flow regime ($Re_{p,mf} < 20$), where the inertia term in the Ergun equation can be safely neglected [13]. This assumption is usually valid for particles in the Geldart group A and C region,

whereas for larger particles, where $Re_{p,mf} > 20$, IPFs are commonly insignificant. There might be cases, however, with a significant level of IPFs, while the condition of the viscous flow regime is violated. These situations include the fluidization of relatively large particles under severe sintering at high temperatures, coated with cohesive materials, or in the presence of liquid bridge forces. The uncertainty in the proposed model caused by neglecting the inertia term under these conditions can undermine the capability of the proposed model to capture the transition between Geldart groups A and C.

Fluidization of nanoparticles: IPFs are at least three to four orders of magnitude greater than W_p for nanoparticles [13]. These powders are, subsequently, located well above the proposed A-C boundary in the Geldart group C region. Nevertheless, nanoparticles can show unique fluidization behaviours, i.e., APF and ABF. On the macroscopic scale, APF and ABF resemble Geldart groups A and B behaviour, respectively. However, the fluidizing entities in APF and ABF are large agglomerates rather than individual particles. Under these conditions, the fluidizing gas mainly flows through intra-agglomerate void spaces [41], while the gas in the void space within agglomerates remains essentially stagnant. Channeling is not observed for these particles since the gas cannot separate or lift the primary nanoparticles even at velocities greater than their U_t . Distinguishing APF and ABF from Geldart group C behaviour remains beyond the scope of this work, while the above discussion provides a framework for future research in the field.

6.5 Conclusion

A model was proposed in this study for the boundary between Geldart groups A and C. It is based on the generalized DHR and U_{mf} deviation equations for the quantification of IPF and the assumption that the A-C boundary corresponds to the conditions where the interstitial gas velocity at U_{mf} equals the particle terminal fall velocity. The first resulting equation correlates the magnitude of IPFs at A/C to ε_{mf} , while the second can be employed to estimate the corresponding agglomerate size. The former was also rearranged in the form of a relationship between d_p , $\rho_p - \rho_g$, IPFs, and ε_{mf} as the generalized form of the original Geldart A-C. This relationship revealed

that, for a given material, d_p at A-C is proportional to the cube root of IPFs. IPFs were subsequently incorporated in the original Geldart chart as a dimension to present the variation of A-C in a 3D plot.

The model showed promising performance in predicting the transition between Geldart groups A and C behaviour. The experimental data from literature suggest that the transition typically happens when IPFs are about an order of magnitude greater than W_p corresponding to the agglomerate to primary particle size ratio of about three. These conditions are met at ε_{mf} in the range of 0.6-0.7. Considering the ratio of W_p at A-B to that at A-C for common powders, the A-B boundary corresponds to the $IPFs/W_p \approx 0.1$.

Acknowledgements

The authors gratefully acknowledge the financial support provided for this research project by the OCP Group and Mitacs through the Mitacs Accelerate program.

6.6 Supplementary Material: Summary of data from literature for the performance evaluation of the proposed A-C

Cases with $IPFs/W_p > 50$ are beyond the upper limit of Y axis in Figure 6.3 (refer to the main manuscript) and, hence, cannot be shown on it. They, however, showed Geldart group C behaviour as expected. The following categories of data were excluded from our analysis:

- 1) Particles with $IPFs/W_p < 0.1$ are categorized as Geldart group B and, hence, were excluded from Table. S1.
- 2) Cases where the available data were not sufficient for adequate quantification of IPFs by the DHR or U_{mf} deviation equations.
- 3) Nanoparticle fluidization.

Table 6.2. Summary of data from literature shown in Figure 6.3 of the main manuscript.

Reference	Solid	d_p	Gas	Temp eratur e	Press ure	$U_{mf,exp}$	$U_{mf,pred}$	ϵ_{mf}	$\frac{IPFs}{W_p}$	Geldart behaviou r	Remarks
		μm		$^{\circ}\text{C}$	kPa	cm/s	cm/s	-	-		
[16]	Iron	3.3	Air	25	101	2.430	0.004	0.615	606.50	C	1- The values of $U_{mf,pred}$ was calculated from the given fluidizability indices and $U_{mf,exp}$. 2- ϵ_{mf} was assumed to be equal to ϵ_0, which was reported in the reference. 3- Cases with quasi-fluidization conditions were categorized as Geldart group C. Geldart group of powders with uncertain fluidization behaviours were determined by Geldart chart as the experiments carried out at ambient conditions with air. Cases with uncertain fluidization
	Nickel	18	Air	25	101	0.530	0.061	0.560	7.63	C	
		81		25	101	3.400	1.600	0.550	1.13	A	
	Coated Nickel	137	Air	25	101	8.300	5.751	0.580	0.44	A	
		137		25	101	10.40	5.751	0.580	0.81	A	
	Glass hollow spheres	35	Air	25	101	10.50	0.034	0.510	307.82	C	
		58		25	101	5.300	0.104	0.540	49.96	C	
		194		25	101	1.250	0.350	0.465	2.57	A	
	Alumina	13	Air	25	101	7.500	0.041	0.630	180.15	C	
		68		25	101	1.300	0.800	0.660	0.63	A	

											behaviour and close to Geldart A-C were excluded.
[1]	Diakon	78	Air	25	101	0.520	0.379	0.520	0.37	A	1- The values of ε_{mf} were estimated as the average of ε_0 and ε_{mb} , whose values were reported in the reference.
		118		25	101	0.820	0.651	0.480	0.26	A	
	Fresh FCC	25	Air	25	101	0.080	0.013	0.569	5.08	A	
		39		25	101	0.090	0.032	0.560	1.82	A	
		55		25	101	0.230	0.097	0.595	1.38	A	
		68		25	101	0.290	0.179	0.608	0.62	A	
		75		25	101	0.330	0.210	0.602	0.57	A	
		100		25	101	0.460	0.378	0.597	0.28	A	
	Spent FCC	45	Air	25	101	0.130	0.057	0.608	1.27	A	
		62		25	101	0.180	0.079	0.565	1.29	A	
		75		25	101	0.220	0.136	0.578	0.67	A	
		87		25	101	0.260	0.154	0.550	0.69	A	

		95		25	101	0.350	0.251	0.575	0.39	A	
[27]	Alumina	88	N ₂	25	100	0.800	0.697	0.653	0.15	A	1- The values of ε_{mf} were estimated as the average of ε_0 and ε_{mb} , whose values were reported in the reference.
		88		25	500	0.870	0.622	0.658	0.40	A	
		88		25	1000	0.830	0.684	0.660	0.21	A	
		88		25	1500	0.870	0.688	0.663	0.26	A	
		88		25	2000	0.900	0.614	0.658	0.47	A	
		88		25	2500	0.880	0.604	0.658	0.46	A	
		88		25	3000	0.890	0.585	0.660	0.52	A	
		88		25	3500	0.890	0.575	0.660	0.55	A	
[28]	sand	84	Air	100	101	0.764	0.689	0.592	0.11	A	
		84		200	101	0.735	0.671	0.615	0.10	A	
		84		300	101	0.667	0.618	0.627	0.08	A	
		84		400	101	0.714	0.587	0.635	0.22	A	
		84		500	101	0.722	0.591	0.640	0.22	A	

		84		600	101	0.844	0.583	0.650	0.45	A	
		103		300	101	1.007	0.855	0.596	0.18	A	
		103		400	101	1.167	0.841	0.608	0.39	A	
		103		500	101	1.251	0.859	0.626	0.46	A	
		103		600	101	1.297	0.895	0.641	0.45	A	
		103		700	101	1.419	0.891	0.649	0.59	A	
		169		100	101	2.653	2.206	0.569	0.20	A	
		169		200	101	2.359	1.994	0.569	0.18	A	
		169		300	101	2.330	1.915	0.587	0.22	A	
		169		400	101	2.301	1.859	0.599	0.24	A	
		169		500	101	2.309	1.836	0.611	0.26	A	
		169		600	101	2.128	1.824	0.625	0.17	A	
		169		700	101	2.326	1.854	0.627	0.25	A	
		209		400	101	4.228	3.688	0.564	0.15	A	

		271		200	101	7.460	6.680	0.543	0.12	A	
		271		300	101	6.977	6.202	0.554	0.13	A	
		271		400	101	6.872	5.906	0.554	0.16	A	
		271		500	101	6.578	5.882	0.572	0.12	A	
		271		700	101	6.406	5.791	0.598	0.11	A	
		271		800	101	6.452	5.856	0.597	0.10	A	
		648		300	101	29.69	25.951	0.510	0.14	A	
		648		400	101	30.23	24.744	0.516	0.22	A	
		648		500	101	29.77	21.563	0.511	0.38	A	
		648		600	101	28.80	20.891	0.517	0.38	A	
		648		700	101	27.16	21.020	0.526	0.29	A	
		648		800	101	29.21	21.298	0.536	0.37	A	
	Glass Ballotini	74	Air	300	101	0.557	0.466	0.468	0.2	A	
		91		100	101	0.768	0.681	0.432	0.13	A	

		91		200	101	0.683	0.607	0.432	0.12	A	
		91		300	101	0.691	0.550	0.451	0.26	A	
		91		400	101	0.752	0.597	0.466	0.26	A	
		156		400	101	2.245	1.714	0.458	0.31	A	
		232		100	101	4.968	4.287	0.446	0.16	A	
		232		200	101	4.576	3.734	0.451	0.23	A	
		232		300	101	3.904	3.304	0.451	0.18	A	
	Salt	601	Air	200	101	30.63	27.251	0.517	0.12	A	
		601		300	101	33.03	26.823	0.533	0.23	A	
		601		400	101	33.60	27.250	0.549	0.23	A	
	FCC	59	Air	100	101	0.446	0.393	0.545	0.14	A	
		59		200	101	0.428	0.339	0.544	0.26	A	
		59		300	101	0.429	0.328	0.553	0.31	A	
		59		400	101	0.431	0.314	0.563	0.37	A	

		59		500	101	0.506	0.303	0.578	0.67	A	
		59		600	101	0.600	0.298	0.581	1.01	A	
[29]	Boron Nitride	8	N ₂	25	0	8.500	0.002	0.420	4071.84	C	1- ε_{mf} was assumed to be equal to ε_0 , which was reported in the reference.
		8		25	2	4.300	0.003	0.475	1288.29	C	
		8		25	9	4.000	0.005	0.520	834.81	C	
		8		25	16	3.200	0.007	0.560	489.76	C	
		8		25	30	2.400	0.007	0.570	340.13	C	
		8		25	35	2.600	0.008	0.580	341.62	C	
		8		25	45	2.600	0.008	0.580	341.64	C	
		8		25	89	2.000	0.008	0.595	234.47	C	
[30]	FCC	76	Air	25	101	0.493	0.432	0.484	0.14	A	
		76		100	101	0.481	0.379	0.494	0.27	A	
		76		200	101	0.477	0.356	0.509	0.34	A	
		76		300	101	0.465	0.341	0.522	0.36	A	

		76		400	101	0.465	0.335	0.536	0.39	A	
		76		500	101	0.493	0.337	0.547	0.46	A	
		76		600	101	0.513	0.331	0.548	0.55	A	
		76		700	101	0.566	0.322	0.563	0.76	A	
		96		200	101	0.579	0.506	0.485	0.14	A	
		96		300	101	0.574	0.471	0.506	0.22	A	
		96		400	101	0.557	0.467	0.520	0.19	A	
		96		500	101	0.565	0.467	0.535	0.21	A	
		96		600	101	0.633	0.458	0.543	0.38	A	
		96		700	101	0.686	0.458	0.565	0.50	A	
	Corundum	67	Air	25	101	0.963	0.831	0.624	0.16	A	
		67		100	101	0.927	0.731	0.628	0.27	A	
		67		200	101	0.871	0.644	0.636	0.35	A	
		67		300	101	0.804	0.623	0.647	0.29	A	

		67		400	101	0.768	0.618	0.651	0.24	A	
		67		500	101	0.801	0.591	0.659	0.36	A	
[31]	Glass beads	6.1	Air	25	101	2.000	0.043	0.750	45.55	C	1- U_{mf} of sound assisted fluidization was considered as $U_{mf,pred}$ for Geldart A particles (individual particle fluidization in virtual absence of IPFs that are broken by sound).
		10		25	101	1.600	0.032	0.720	49.14	C	
		39		25	101	0.320	0.100	0.540	2.20	A	
		65		25	101	0.550	0.200	0.600	1.75	A	
	FCC	65	Air	25	101	0.300	0.100	0.550	2.00	A	
	TiO ₂	5.2	Air	25	101	1.600	0.062	0.868	24.87	C	
[32]	Cornstarch	15	N ₂	25	101	1.400	0.123	0.700	10.35	C	1- Cases with more than 0.1 wt% nanoparticle addition (corresponding to greater than 10% surface coverage) were excluded.
		15		25	101	0.750	0.123	0.700	5.08	C	
		15		25	101	0.720	0.123	0.700	4.84	C	
		15		25	101	0.400	0.123	0.700	2.24	A	
		15		25	101	0.350	0.123	0.700	1.84	A	
		207	Air	40	101	7.000	2.155	0.520	2.25	A	

[33]	Placebo granule	207		40	101	7.200	2.155	0.540	2.34	A	
		207		40	101	15.00	2.155	0.620	5.96	A	
		207		40	101	25.00	2.155	0.680	10.60	C	
		207		40	101	30.00	2.155	0.690	12.92	C	
		207		40	101	40.00	2.155	0.700	17.56	C	
[34]	Iron	90	H ₂	100	101	10.50	9.000	0.700	0.17	A	1- The values of $U_{mf,pred}$ and $U_{mf,exp}$ were presented in the reference. The values of $U_{mf,pred}$ presented in the reference were slightly adjusted such that the prediction fits the experimental data at ambient conditions, where the particles are expected to behave like Geldart group B. 2- The values of ε_{mf} were available at ambient conditions only. Similar values were considered for higher temperatures.
		90		210	101	10.00	7.300	0.700	0.37	A	
		90		320	101	11.00	6.000	0.700	0.83	A	
		90		370	101	12.00	5.600	0.700	1.14	A	
		90		425	101	13.00	5.300	0.700	1.45	A	
		90	N ₂	100	101	5.200	4.700	0.700	0.11	A	
		90		210	101	4.900	4.000	0.700	0.23	A	
		90		320	101	4.800	3.600	0.700	0.33	A	
		90		370	101	4.700	3.300	0.700	0.42	A	

		90		425	101	4.900	3.000	0.700	0.63	A	
		90		475	101	5.000	2.800	0.700	0.79	A	
		90		530	101	6.500	2.600	0.700	1.50	A	
		90	Argon	475	101	3.000	2.100	0.700	0.43	A	
		90		530	101	4.000	2.000	0.700	1.00	A	
		90		580	101	5.000	1.900	0.700	1.63	A	
[10]	Coated sugar beads	580	Air	20	101	18.00	15.960	0.490	0.13	A	1- The value of $U_{mf,exp}$ for uncoated particle at 20°C (Geldart group B) was considered as $U_{mf,pred}$ at this temperature. It was then adjusted for 30°C and 40°C considering the difference in the gas viscosity and density between the two temperatures.
		580		30	101	25.00	15.542	0.500	0.61	A	
		580		40	101	40.00	15.151	0.530	1.64	A	
[25]	Silica sand	60	N ₂	20	101	0.670	0.386	0.550	0.74	A	1- Data of sound assisted fluidization under 50 dB and 150 Hz were used for ash particles,
		60		200	101	0.680	0.317	0.570	1.14	A	
		60		400	101	0.750	0.270	0.590	1.78	A	

		60		600	101	0.810	0.246	0.610	2.29	A	which could not be normally fluidized.
	Ash	8	N ₂	20	101	0.067	0.006	0.605	11.07	C	
		8		100	101	0.071	0.005	0.610	13.84	C	
		8		200	101	0.079	0.004	0.620	17.14	C	
		8		300	101	0.085	0.004	0.640	18.15	C	
		8		400	101	0.096	0.004	0.630	24.99	C	
		8		500	101	0.115	0.004	0.650	28.34	C	
		8		800	101	0.128	0.003	0.650	47.65	C	
[35]	Glass beads	39	Air	25	101	0.700	0.239	0.550	1.93	A	
[37]	Glass beads	39	Air	25	101	0.500	0.200	0.550	1.50	A	1- ε_0 and ε_{mf} were estimated based on the values for similar particles in our experiments [7]. 2- $U_{mf,pred}$ for pure materials were estimated assuming $IPFs/W_p$ to be about 1.5 and 0.5 for 39-
	Glass beads + additive	39		25	101	2.100	0.200	0.570	9.50	A	
		39		25	101	3.100	0.200	0.590	14.50	C	
		39		25	101	3.700	0.200	0.610	17.50	C	
	FCC	76	Air	25	101	0.160	0.105	0.550	0.52	A	

	FCC + additive	76		25	101	0.220	0.105	0.570	1.10	A	micron glass beads and 76-micron FCC [7, 13]. 3- $U_{mf,pred}$ for cases with additives were estimated considering the effect of additive on the average d_p and ρ_p .
		76		25	101	1.000	0.105	0.590	8.52	C	
		76		25	101	1.750	0.105	0.610	15.67	C	
[7]	Glass beads	67	Air	20	101	0.692	0.520	0.479	0.33	A	
		67		90	101	0.568	0.447	0.474	0.27	A	
		67		115	101	0.629	0.452	0.487	0.39	A	
		67		150	101	0.830	0.460	0.538	0.80	A	
		67		200	101	0.991	0.475	0.579	1.09	A	
	FCC	83	Air	20	101	0.565	0.355	0.453	0.59	A	
		83		110	101	0.434	0.274	0.436	0.58	A	
		83		200	101	0.446	0.245	0.449	0.82	A	
		83		300	101	0.476	0.233	0.470	1.04	A	
		83		400	101	0.480	0.210	0.475	1.29	A	

		83		500	101	0.583	0.181	0.487	2.21	A	
		83		600	101	0.617	0.162	0.490	2.82	A	
	Glass beads	67	Argon	20	101	0.580	0.450	0.477	0.29	A	
		67		90	101	0.454	0.360	0.466	0.26	A	
		67		115	101	0.516	0.363	0.491	0.42	A	
		67		150	101	0.686	0.371	0.549	0.85	A	
		67		200	101	0.736	0.363	0.566	1.03	A	
	FCC	83	Argon	20	101	0.448	0.290	0.452	0.55	A	
		83		110	101	0.331	0.221	0.440	0.50	A	
		83		200	101	0.330	0.195	0.446	0.69	A	
		83		300	101	0.356	0.185	0.474	0.93	A	
		83		400	101	0.363	0.165	0.465	1.20	A	
		83		500	101	0.435	0.142	0.484	2.06	A	
		83		600	101	0.499	0.126	0.492	2.95	A	

[36]	Corundum	45	Air	25	101	0.590	0.512	0.620	0.15	A	
		45		100	101	0.550	0.453	0.627	0.21	A	
		45		200	101	0.530	0.400	0.634	0.32	A	
		45		300	101	0.500	0.368	0.642	0.36	A	
		45		400	101	0.480	0.347	0.649	0.38	A	
		45		500	101	0.440	0.333	0.656	0.32	A	
		45		600	101	0.430	0.324	0.663	0.33	A	
		45		700	101	0.420	0.318	0.670	0.32	A	
[13]	Iron oxide (Fe ₃ O ₄)	0.5	Air	20	101	5.040	0.002	0.881	2563.77	C	
	Iron oxide (Fe ₂ O ₃)	1		20	101	4.450	0.005	0.844	843.30	C	
	Silicon oxide (SiO ₂)	3		20	101	3.590	0.020	0.813	181.63	C	
	Calcium oxide (CaO)	6		20	101	3.520	0.081	0.795	42.34	C	

	LFP (LiFePO ₄)	14		20	101	4.890	0.284	0.723	16.21	C	
	Cornstarch	18		20	101	2.890	0.177	0.714	15.37	C	
	Glass beads	20		20	101	0.546	0.129	0.618	3.22	A	
	Wheat flour	26		20	101	3.710	0.284	0.685	12.06	C	
	Sugar	31		20	101	6.345	0.491	0.697	11.92	C	
	Phosphogyp sum (CaSO ₄ ·2H ₂ O)	35		20	101	1.510	0.259	0.646	4.84	A	

6.7 Nomenclature

Acronyms

ABF	agglomerating bubbling fluidization
A-B	Geldart groups A and B boundary
A-C	Geldart groups A and C boundary
AOR	angle of repose
APF	agglomerating particulate fluidization
DHR	dynamic Hausner ratio
FCC	fluid cracking catalyst
HR	Hausner ratio
IPFs	interparticle forces

Symbols

Ar	Archimedes number (-)
a_s	separation distance between neighboring particles at the contact point (m)
C	constant in the Carman–Kozeny equation (-)
$d_{agg,mf}$	effective agglomerate diameter at minimum fluidization conditions (m)
$\frac{d_{agg,mf}}{d_p} \Big _{A-C}$	agglomerate size ratio at A-C boundary (-)
d_p	particle diameter (m)
$d_p \Big _{A-C}$	particle diameter at A-C boundary (m)

$d_p _{A-C, ambient}$	particle diameter at the A-C boundary at ambient conditions (m)
F_D	drag force acting on a particle (N)
F_{vdW}	pairwise van der Waals forces between two neighboring particles (N)
g	gravitational acceleration (m/s ²)
H	Hamaker constant (J)
$IPFs$	interparticle forces acting on each particle (N)
$IPFs_{amb}$	interparticle forces acting on each particle at ambient conditions (N)
$\frac{IPFs}{W_p} _{A-C}$	ratio of interparticle forces and weight of a particle at the A-C boundary (N)
$\frac{IPFs}{W_p} _{A-B}$	ratio of interparticle forces and weight of a particle at the A-B boundary (N)
k	coordination number (-)
r	asperity size (m)
$Re_{p,mf}$	particle Reynolds number at minimum fluidization conditions (-)
$\hat{S}_e$	specific surface area index (m ³ /kg)
U_g	superficial gas velocity (m/s)
U_{mf}	minimum fluidization velocity (m/s)
$U_{mf,exp}$	experimental minimum fluidization velocity (m/s)
$U_{mf,pred}$	minimum fluidization velocity predicted by the Carman-Kozeny equation (m/s)
U_t	terminal fall velocity of a particle (m/s)
W_p	weight of a single particle (N)

Greek letters

ε_0	loose bulk voidage (-)
ε_{mf}	minimum fluidization voidage (-)
μ_g	gas viscosity (Pa. s)
ρ_g	gas density (kg/m ³)
ρ_p	particle density (kg/m ³)

CHAPTER 7 QUANTIFICATION OF AGGLOMERATE SIZE AND MAGNITUDE OF INTERPARTICLE FORCES WITH MODIFIED HAUSNER RATIO

7.1 Introduction

Interparticle forces (IPFs) play a crucial role in the handling and processing of particulate materials. These forces, such as van der Waals, electrostatic, and material bridge forces, determine the interactions between particles and significantly influence various aspects of their behaviour. Hence, understanding and manipulating IPFs are essential for achieving desired outcomes in the processes involving particulate materials. Moreover, in fluidization, IPFs can affect the bed hydrodynamics, i.e., the patterns of the movement of suspended particles and the gas flow. Consequently, they impact the heat and mass transfer within the system and, ultimately, the performance of the fluidized bed. By controlling IPFs, fluidization processes can be optimized to ensure efficient solid mixing, enhanced gas-solid contact, and improved heat and mass transfer, leading to better performance and productivity in various industrial applications of these process units.

Due to their unique properties and versatile applications, fine and ultrafine powders hold immense significance in chemical, pharmaceutical, and material industries. One primary advantage of these powders is their high surface area per unit mass, which allows for enhanced gas-solid contact. This high surface area is particularly advantageous in catalysis, where fine and ultrafine powders can provide greater catalytic activity, leading to improved chemical reactions and process efficiency. These powders, however, often exhibit unfavorable characteristics of Geldart [1] Group C due to the substantial magnitude of IPFs that govern their fluidization behaviour [2, 3].

IPFs influence the fluidization behaviour of a particulate assembly via two primary pathways. Firstly, these forces contribute to an increase in the bed voidage [4-7], enhancing the contact between gas and solid particles [8]. The second pathway is the formation of agglomerates that reduces the available surface area for gas-solid contact [3]. This decrease in the gas-solid contact can undermine the advantage of fine powders in providing a high surface area. This effect is particularly problematic in beds containing ultrafine particles and nanopowders, as they exhibit an

enhanced gas screening effect [9]. Gas screening refers to the primary passage of the gas through the void spaces between agglomerates without efficient contact with the primary particles within the agglomerates.

Hausner ratio (HR) is defined as the ratio of loose and tapped bulk densities:

$$HR = \frac{\rho_{tap}}{\rho_0} = \frac{1 - \varepsilon_{tap}}{1 - \varepsilon_0} \quad 7.1$$

where ρ_{tap} and ρ_0 represent the bulk density in the tapped and loose states, respectively, while ε_{tap} , and ε_0 refer to the voidage in these states.

HR is a qualitative index that provides insight into the magnitude of IPFs acting on particles in a particulate assembly. A low HR, i.e., HR close to one, indicates that the particles have good flowability, suggesting weak interparticle forces. On the contrary, higher HR values correspond to greater magnitudes of IPFs. For instance, particles with HR greater than 1.4 were suggested to show Geldart group C behaviour [10]. In contrast, HR smaller than 1.25 correspond to Geldart groups A, B, or D behaviours [10]. The advantage of the HR lies in its simplicity and ease of calculation, as it merely involves the measurement of the powder's bulk density under two conditions. However, HR cannot be employed to quantify IPFs since no relationship between these forces and HR has yet been developed.

This study presents a model for the relationship between bed voidage and agglomerate size. The model leads to two relationships correlating agglomerate size and magnitude of IPFs to the voidage of a particulate assembly. For particles with Geldart group C behaviour, these relationships are formulated as correlations between agglomerate size and magnitude of IPFs with HR.

7.2 Model development

7.2.1 Background and Assumptions

At ambient conditions and in a dry environment, van der Waals forces are the dominant type of IPFs [11-13]. This type of IPFs is more of a function of the particle's surface properties rather than its size [11-13]. Hence, The magnitude of IPFs at ambient conditions, i.e., the ratio of IPFs acting on a particle to its weight ($IPFs/W_p$) increases with decreasing particle diameter, d_p , due to the proportionality of W_p with d_p^3 . This variation of the magnitude of IPFs is reflected in the correspondence of different Geldart behaviours with varying particle diameter ranges. For instance, the cohesive behaviour of Geldart group C is typically observed for particles with $d_p < 20 \mu m$ at ambient conditions due to the dominance of IPFs over W_p for this range of particle diameter ($IPFs/W_p > \sim 10$ [14]). On the other hand, IPFs are negligible in comparison with W_p ($IPFs/W_p < \sim 0.1$ [14]) for Geldart groups B – and D, which approximately correspond to $d_p > 100 \mu m$ at ambient conditions. Between the above two classes, i.e., for $\sim 20 < d_p < \sim 100 \mu m$, there exists Geldart group A where the IPFs and W_p are comparable ($\sim 0.1 < IPFs/W_p < \sim 10$ [14]).

The voidage of a particulate assembly, including loose (ε_0), tapped (ε_{tap}), and minimum fluidization (ε_{mf}) voidages, increases with a growth in the magnitude of IPFs [4-7] and, hence, with decreasing d_p . We will show in Section 7.4.1 that this growth cannot be directly attributed to the decrease in d_p . This remark is supported by relatively constant ε_0 , ε_{mf} , and ε_{tap} with varying particle diameter in the Geldart group B region (see Figure 7.1). By decreasing d_p in Geldart group A, the bed voidage in different states begin to increase. This initial increase can be explained by the enhanced non-uniformity of the packing structure caused by IPFs. Under these conditions, small bridges can develop that are strong enough to withstand the weight of the particles [15], creating small voids that contribute to bed voidage. Agglomerates form and function as secondary porous particles by increasing the magnitude of IPFs for smaller particles. The contribution of the porosity of the agglomerates to the bed voidage is the main mechanism for the sharper increase in

ε_0 and ε_{tap} for finer Geldart group A and Geldart group C powders (see Figure 7.1). The voidage of a particulate assembly under these conditions, therefore, can be estimated by the sum of the contributions of the voidage of agglomerates, ε_{agg} , and intra-agglomerate voidage, ε_{i-agg} .

Electrostatic and van der Waals forces vanish in water [12, 16] due to its high dielectric constant. Accordingly, the water phase fraction in water-solid systems, ε_w , corresponds to the behaviour of particles with virtually no IPFs [7]. Therefore, by comparing ε_0 and ε_w , one can gain insight into the presence or absence of IPFs. The absence of IPFs corresponds to the conditions where ε_0 and ε_w are essentially similar, whereas the presence of these forces causes an increase in ε_0 leading to the conditions where ε_0 is greater than ε_w . The former condition happens for Geldart groups B and D, while the latter is the case for groups A and C. As alluded to earlier, ε_0 for Geldart groups B and D is independent of d_p while varying in the range of about 0.35-0.55 depending on the particles' shape. Therefore, ε_w , which is nearly equal to ε_0 of these particles (see Figure 7.1), is also a function of particles' shape and varies in a similar range.

Large agglomerates are composed of numerous particles. Only a few of these particles are in contact with the outer layer of adjacent agglomerates. As a result, inter-agglomerate forces (IAFs) must be approximately equal to the IPFs between individual particles [17]. The weight of an agglomerate (W_{agg}), on the other hand, is substantially greater than that of a single particle. These remarks lead to the conclusion that the ratio of $IAFs/W_{agg}$ is small for large agglomerates. Consequently, the packing structure of these secondary particles is likely to resemble that of large particles with negligible magnitudes of IPFs. Therefore, intra-agglomerate voidage can be approximated by ε_0 ($\approx \varepsilon_w$) of large particles of Geldart group B or D with a similar shape as the agglomerates. ε_{i-agg} is not necessarily similar to ε_0 or ε_w of Geldart groups B and D particles of the same material as agglomerates might have a different shape than these particles. Nevertheless, ε_{i-agg} must have a value in the approximate range of 0.35-0.55 since ε_0 or ε_w of Geldart groups B and D particles with all kinds of shpes vary in this range.

In the next section, we derive a relationship between agglomerate size and voidage of a particulate assembly. This relationship will eventually help quantify the magnitude of IPFs. Based on the above discussions, we adopted the following assumptions for the development of this model:

- (i) Particles are identical.
- (ii) Voidage of a powder with Geldart group C behaviour is the sum of the contributions of ε_{agg} and ε_{i-agg} .
- (iii) Inter-agglomerate forces acting on an agglomerate are negligible in comparison with W_{agg} . Therefore, agglomerates are assumed to mimic the packing structure of large particles with a negligible magnitude of IPFs.
- (iv) Following assumption (iii), ε_{i-agg} is assumed to be equal to ε_w of particles of Geldart groups B and D with a similar shape as the agglomerates.
- (v) $\varepsilon_{mf} \approx \varepsilon_0$ for Geldart group C behaviour due to the occurrence of channeling at minimum fluidization for this Geldart class.
- (vi) The generalized DHR equation [3] is employed to quantify the magnitude of IPFs.

7.2.2 Development of Modified Hausner ratio model

We recently introduced the generalized form of the DHR (dynamic Hausner ratio, i.e., the ratio of $\varepsilon_{mf}/\varepsilon_0$) equation [3] to quantify the magnitude of IPFs acting on a particle in a gas-solid fluidized bed. This equation is a relationship between $IPFs/W_p$ and the effects of IPFs on the particulate assembly at minimum fluidization, i.e., an increase in the bed voidage, which is reflected in $\varepsilon_{mf}/\varepsilon_0$, and the formation of larger agglomerates, which is signified by $d_{agg,mf}/d_p$:

$$\frac{IPFs}{W_p} = \left(\frac{d_{agg,mf}}{d_p} \right)^2 \left(\frac{\varepsilon_{mf}}{\varepsilon_0} \right)^3 \left(\frac{1 - \varepsilon_0}{1 - \varepsilon_{mf}} \right) - 1 \quad 7.2$$

where $d_{agg,mf}$ is the agglomerate size at minimum fluidization. For Geldart group C behaviour, ε_{mf} is equal to ε_0 based on the assumption (v), leading to a simplified version of the DHR equation for these powders:

$$\frac{IPFs}{W_p} = \left(\frac{d_{agg,mf}}{d_p} \right)^2 - 1 \quad 7.3$$

Hence, we can quantify the magnitude of IPFs if $d_{agg,mf}/d_p$ is known. In the following, a model is developed to correlate this ratio to the bed voidage.

Following assumption (ii), ε_{mf} of a particulate assembly can be expressed as:

$$\varepsilon_{mf} = \varepsilon_{i-agg,mf} + \varepsilon_{agg,mf}(1 - \varepsilon_{i-agg,mf}) \quad 7.4$$

where $\varepsilon_{agg,mf}$ and $\varepsilon_{i-agg,mf}$ refer to inter-agglomerate and intra-agglomerate voidages at minimum fluidization conditions. Therefore, the first and second terms on the right-hand side of Eq. 7.2 correspond to the contributions of intra-agglomerate and inter-agglomerate voidages, respectively.

$\varepsilon_{agg,mf}$ can be expressed by:

$$\varepsilon_{agg,mf} = 1 - \frac{N_{p,agg} V_p}{V_{agg,mf}} \quad 7.5$$

where $N_{p,agg}$ is the number of particles in an agglomerate. V_p and $V_{agg,mf}$ are the volume of a particle and an agglomerate at minimum fluidization, respectively:

$$V_p = \frac{\pi}{6} d_p^3 \quad 7.6$$

$$V_{agg,mf} = \frac{\pi}{6} d_{agg,mf}^3 \quad 7.7$$

The fractal dimension of an agglomerate D_a is defined as [12]:

$$D_a = \frac{\ln N_{p,agg}}{\ln \left(\frac{d_{agg}}{d_p} \right)} \quad 7.8$$

Therefore, at minimum fluidization conditions:

$$N_{p,agg} = \left(\frac{d_{agg,mf}}{d_p} \right)^{D_a} \quad 7.9$$

Replacing Eqs. 7.6, 7.7, and 7.9 in Eq. 7.5 results in:

$$\varepsilon_{agg,mf} = 1 - \left(\frac{d_p}{d_{agg,mf}} \right)^{3-D_a} \quad 7.10$$

By substitution of Eq. 7.10 for $\varepsilon_{agg,mf}$ in Eq. 7.4, we obtain:

$$\varepsilon_{mf} = \varepsilon_{i-agg,mf} + \left[1 - \left(\frac{d_p}{d_{agg,mf}} \right)^{3-D_a} \right] (1 - \varepsilon_{i-agg,mf}) \quad 7.11$$

$d_{agg,mf}/d_p$ can be calculated as a function of ε_{mf} and $\varepsilon_{i-agg,mf}$ by Eq. 7.11, as follows:

$$\frac{d_{agg,mf}}{d_p} = \left(\frac{1 - \varepsilon_{i-agg,mf}}{1 - \varepsilon_{mf}} \right)^{\left(\frac{1}{3-D_a} \right)} \quad 7.12$$

Eq. 7.12 is a relationship between the agglomerate size, fractal dimension of agglomerates, and the ratio of $\frac{1-\varepsilon_{i-agg,mf}}{1-\varepsilon_{mf}}$, which we called modified Hausner ratio (MHR). The reason behind this terminology will be discussed in Section 7.2.3.3. Hence, Eq. 7.12 can be written as follows:

$$\frac{d_{agg,mf}}{d_p} = MHR^{\left(\frac{1}{3-D_a} \right)} \quad 7.13$$

Following the estimation of $d_{agg,mf}/d_p$ by Eq. 7.13, the simplified DHR equation, Eq. 7.3, can be employed to quantify the magnitude of IPFs. By replacing $d_{agg,mf}/d_p$ from Eq. 7.13 in Eq. 7.3, we obtain:

$$\frac{IPFs}{W_p} = MHR^{\left(\frac{2}{3-D_a} \right)} - 1 \quad 7.14$$

Eqs. 7.13 and 7.14 are the general form of MHR equations as relationships between $d_{agg,mf}/d_p$ and $IPFs/W_p$ with MHR and fractal dimension of the agglomerates. For micron-size particles, however, D_a has been reported to have a robust value of 2.5 [12], leading to simplified versions of the MHR equations:

$$\frac{d_{agg,mf}}{d_p} = MHR^2 \quad 7.15$$

$$\frac{IPFs}{W_p} = MHR^4 - 1 \quad 7.16$$

The simplified MHR equations can quantify the magnitude of IPFs and agglomerate size once the value of MHR is known. In the next section, we present practical equations for estimating MHR.

7.2.3 Modified Hausner ratio

In this section, we discuss the calculation of MHR for particles with different Geldart group behaviours. At the end of this section, a comparison between HR and MHR will be presented to justify the adopted terminology for the MHR.

7.2.3.1 Individual particle fluidization

As alluded to earlier, MHR is defined as:

$$MHR = \frac{1 - \varepsilon_{i-agg,mf}}{1 - \varepsilon_{mf}} \quad 7.17$$

For individual particle fluidization, $\varepsilon_{i-agg,mf}$ can be replaced by ε_w :

$$MHR = \frac{1 - \varepsilon_w}{1 - \varepsilon_{mf}} \quad 7.18$$

Eq. 7.18 can be employed to estimate MHR under individual particle fluidization conditions, which is the case for larger particles with Geldart group A behaviour and groups B and D behaviours [7]. For Geldart groups B and D, ε_w and ε_{mf} are similar to ε_0 (see Figure 7.1) leading to $MHR \approx 1$. This results in the estimation of $d_{agg,mf}/d_p \approx 1$ and $IPFs/W_p \approx 0$ by Eqs. 7.15 and 7.16 for these particles. Hence, the results of the MHR equations for groups B and D behaviours are in accordance with our expectations, even though these equations were originally developed for agglomerating conditions.

7.2.3.2 Agglomerating fluidization

In Section 7.2.2, we estimated the voidage of an agglomerate by its fractal dimension, i.e., Eq. 7.10. $\varepsilon_{agg,mf}$ can also be estimated based on ε_{tap} with the following equation [3]:

$$\varepsilon_{agg,mf} = \varepsilon_{tap} \left[1 - \left(\frac{d_p}{d_{agg,mf}} \right)^3 \right] \quad 7.19$$

Equating the right-hand sides of Eqs. 7.10 and 7.19 results in:

$$1 - \left(\frac{d_p}{d_{agg,mf}} \right)^{3-D_a} - \varepsilon_{tap} + \varepsilon_{tap} \left(\frac{d_p}{d_{agg,mf}} \right)^3 = 0 \quad 7.20$$

In comparison with ε_{tap} , the term $\varepsilon_{tap} \left(\frac{d_p}{d_{agg,mf}} \right)^3$ can be safely neglected for Geldart group C behaviour where $d_{agg,mf}/d_p > \sim 3$. Subsequently, Eq. 7.20 can be solved for $d_{agg,mf}/d_p$ as follows:

$$\frac{d_{agg,mf}}{d_p} = \left(\frac{1}{1 - \varepsilon_{tap}} \right)^{\left(\frac{1}{3-D_a} \right)} \quad 7.21$$

Comparison of Eqs. 7.13 and 7.21 reveals that:

$$MHR = \frac{1}{1 - \varepsilon_{tap}} \quad 7.22$$

Eq. 7.22 implies that MHR is the ratio of particle density to the tapped bulk density:

$$MHR = \frac{\rho_p}{\rho_{tap}} \quad 7.23$$

Eq. 7.22 can also be reformulated as:

$$MHR = \frac{1}{HR(1 - \varepsilon_0)} \quad 7.24$$

Either of the above three equations can be employed to estimate the value of MHR under agglomerating conditions, i.e., for Geldart group C powders and finer particles with Geldart group A behaviour.

$\varepsilon_{i,agg,mf}$ can also be calculated by replacing MHR in Eq. 7.22:

$$\frac{1 - \varepsilon_{i-agg,mf}}{1 - \varepsilon_{mf}} = \frac{1}{1 - \varepsilon_{tap}} \quad 7.25$$

Therefore:

$$\varepsilon_{i-agg,mf} = 1 - \frac{1 - \varepsilon_{mf}}{1 - \varepsilon_{tap}} \quad 7.26$$

As $\varepsilon_{mf} \approx \varepsilon_0$ for Geldart group C behaviour, Eq. 7.26 can be expressed as:

$$\varepsilon_{i-agg,mf} = 1 - \frac{1}{HR} \quad 7.27$$

Eq. 7.27 can be employed to estimate $\varepsilon_{i-agg,mf}$ by solely measuring HR for Geldart group C powders. As discussed before, the value of $\varepsilon_{i-agg,mf}$ must be in the range of about 0.35-0.55 based on the shape of the agglomerates. HR values were reported to vary in the range of about 1.4-2.2

for micron-size Geldart group C powders [10]. These values result in the estimation of $\varepsilon_{i-agg,mf}$ in the range of $\sim 0.29 - 0.55$, which is remarkably close to our range of expectations.

7.2.3.3 Comparison of MHR and HR

Below, we present a comparison between HR and MHR to evaluate their relationships with IPFs:

- Both HR and MHR are ratios of different solid phase fractions. HR is the ratio of solid phase fractions of the bulk in the tapped and loose conditions. MHR can be viewed as the ratio of solid phase fractions of the fluidizing entities in the absence and presence of IPFs based on Eq. 7.22. According to this equation, MHR is equal to one divided by the solid phase fraction of the tapped bulk. In the absence of IPFs, primary particles, with a solid phase fraction equal to one, are the fluidizing entities, whereas agglomerates with a solid phase fraction approximately equal to $1 - \varepsilon_{tap}$ are the fluidizing entities in the presence of IPFs.
- Both HR and MHR are ratios of different densities. HR is the ratio of bulk densities in tapped and loose states. MHR is the ratio of particle density to the tapped bulk density based on Eq. 7.23. Similar to the previous remark, MHR can also be considered as the ratio of the densities of fluidizing entities in the absence and presence of IPFs. $d_{agg,mf}$ is equal to d_p in the absence of IPFs, implying that ρ_p is the density of the fluidizing entities under these circumstances. In the presence of IPFs, on the other hand, agglomerates with a density close to ρ_{tap} [3] are the fluidizing entities.
- Even for particles with an insignificant magnitude of IPFs, the HR is still slightly greater than one (see Figure 7.2). When a particle assembly is tapped or subjected to external stress, particles rearrange and pack more efficiently, reducing the void space between particles. This enhanced packing structure of the tapped bulk results in a Modestly higher bulk density and HR slightly greater than one. Therefore, the deviation of HR from one is not exclusively because of IPFs. MHR, on the other hand, is very close to one if IPFs are negligible, i.e.,

for particles with Geldart groups B and D behaviour (see Figure 7.2). Considering the above remarks, the deviation of MHR from one is entirely due to IPFs.

- HR is qualitatively related to IPFs, while MHR helps quantify IPFs by Eqs. 7.14 or 7.16. MHR is also correlated with the agglomerate size by Eqs. 7.13 or 7.15.

The above remarks explain the terminology employed in this work for MHR as the modified version of the Hausner ratio.

7.3 Experimental

The experiments were performed with four solid materials with several particle sizes. They include fluid cracking catalyst (FCC), glass beads (GB), silica sand (SS), and Silicon carbide (SC). Particle density was measured with a micromeritics® AccuPyc II 1340 Pycnometer with Helium as the penetrating gas. Particle size was measured by the wet mode analysis of EyeTech particle size and shape analyzer. In the wet mode, the particles are poured into water to minimize the van der Waals forces and break the agglomerates [3]. Table 1 presents the physical characteristics of the investigated powders. Sieving information shows relatively narrow size distributions, which is consistent with the assumption of uniformly sized particles adopted for the development of the MHR equations.

Table 7.1. Physical characteristics of the particles under investigation.

Material	Particle density (kg/m ³)	Sieving size (μm)	Sauter mean particle size (μm)	$U_{mf,exp}$ (cm/s)	Shape factor (-)	d_p (μm)	Geldart group behaviour	Acronym
Fluid Cracking catalyst	1460	180-300	232	3.04	0.94	218	B	FCC-218
		150-180	162	1.43		152	B	FCC-152
		125-150	135	0.86		127	B	FCC-127

		90-125	104	0.87		98	A	FCC-98
		63-90	72	0.56		68	A	FCC-68
		45-63	51	0.49		48	A	FCC-48
		0-45	21	0.52		20	C	FCC-20
		0-25	12	NF**		11	C	FCC-11
Glass beads (GB)	2588	300-430	351	- *	0.96	337	B	GB-337
		125-180	144	2.32		138	B	GB-138
		90-125	109	1.27		105	B	GB-105
		53-90	68	0.84		65	A	GB-65
		32-53	41	0.66		39	A	GB-39
		0-32	20	0.54		19	A	GB-19
		0-12	8	0.61		8	C	GB-8
Silica sand (SS)	2650	300-600	374	- *	0.69	258	B	SS-258
		180-300	227	4.83		157	B	SS-157
		106-180	132	2.1		91	A	SS-91
		75-106	88	1.32		61	A	SS-61
		53-75	62	1.12		43	A	SS-43
		32-53	42	1.44		29	A	SS-29
		0-32	17	NF**		12	C	SS-12
		0.5-10	4	3.59		3	C	SS-3

Silicon carbide (SC)	3071	150-180	166	4.4	0.66	110	B	SC-110
		106-150	122	2.44		81	B	SC-81
		80-106	90	1.62		59	A	SC-59
		53-80	68	1.25		45	A	SC-45
		38-53	44	0.8		29	A	SC-29
		10-38	22	0.74		15	A	SC-15
		1-15	6	0.86		4	C	SC-4
		0-5	2	NF**		1	C	SC-1

* $U_{mf,exp}$ was greater than the range of the employed rotameters and, hence, could not be reliably measured.

** Not fluidizable.

Fluidization experiments were conducted under ambient conditions using a transparent Plexiglas fluidized bed with a diameter of 5.25 cm. To ensure uniform air distribution across the bed, a porous distributor plate was employed, and the windbox was filled with 5 mm glass beads. The fluidized bed apparatus was electrically grounded to prevent the accumulation of electrostatic charges, which could impact the powders behaviour over time. To precisely control the gas flow rate within a broad range of superficial gas velocities, three calibrated rotameters were installed on the air inlet line.

Experimental minimum fluidization velocity, $U_{mf,exp}$, was measured according to Richardson's classical method [18] based on the pressure drop profile obtained during the gradual reduction of gas velocity. In this method, $U_{mf,exp}$ is defined as the superficial gas velocity where the linearly varying bed pressure drop in the fixed bed state intersects with the nearly constant pressure drop at the fully fluidized state. The corresponding bed voidage was considered as ε_{mf} . The bed voidage at complete shutdown of the gas flow was considered as ε_0 .

The particle shape factor was measured with the pressure drop data in the fixed bed state according to the Ergun equation [19], as suggested by Kunii and Levenspiel [20]. The shape factors measured by this approach were consistent with those from the EyeTech particle size and shape analyzer, within a 9% difference. As all particle sizes of a given material have similar shapes, their shape factors were measured for one particle, i.e., the largest particle of each material. The magnitude of IPFs is insignificant for these particles that showed Geldart group B behaviour. Employing shape factors estimated by pressure drop data for these particles guarantees that their experimental and theoretical minimum fluidization velocities become equal. These values result in the magnitude of IPFs being equal to zero – based on U_{mf} deviation equation [3] – for these particles and, therefore, reported in Table 1. The effective particle diameter, d_p , which is referred to as the particle diameter for convenience, was determined by multiplying the average particle size by its corresponding shape factor [20].

ε_{tap} was measured by manual tapping of the bed for about 60 seconds, after which the bed voidage remained steady. The procedure proposed by Soleimani et al. [7] was adopted to measure the water phase fraction in water-solid systems. A graduated cylinder with a diameter of 5.25 cm was equipped with a porous plate at the bottom. The dry powders were carefully poured into the cylinder in their loose bulk condition. To minimize destruction to the structure of the particles bulk, water was gently introduced into the cylinder from the porous plate at the bottom. This allowed the water to fill the voids between particles while preserving the overall structure of the particulate assembly. ε_w was calculated by visually observing the height of the particulate assembly in the graduated cylinder.

7.4 Results and discussion

7.4.1 Variations of ε_0 , ε_{mf} , ε_{tap} , and ε_w with particle diameter

Experimental values of ε_0 , ε_{tap} , ε_{mf} , and ε_w for particles under investigation are presented in Figure 7.1. The corresponding values of HR and MHR are shown in Figure 7.2. The values of

MHR were estimated by Eq. 7.18 for particles with Geldart groups B and A and by Eq. 7.22 for Geldart group C powders. Figure 7.1 and Figure 7.2 represent the behaviour of particulate materials with relatively narrow size distributions (see Table 1). The wide size distribution of particles can lead to additional complexities beyond this study's scope. Due to the high viscosity of water, it cannot easily flow between fine particles. Hence, ε_w could not be adequately measured for particles smaller than about $50 \mu\text{m}$ by the procedure described in Section 7.3.

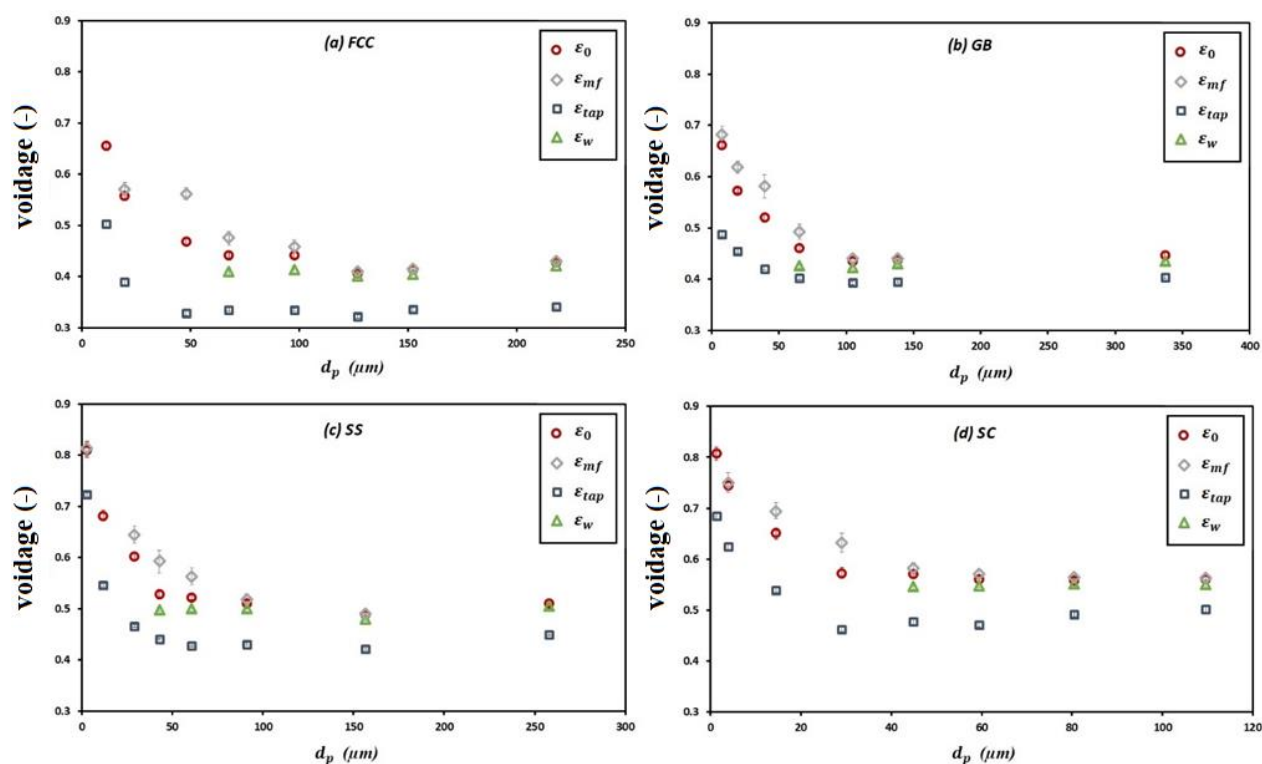


Figure 7.1. Variations of ε_0 , ε_{tap} , ε_{mf} , and ε_w with particle diameter.

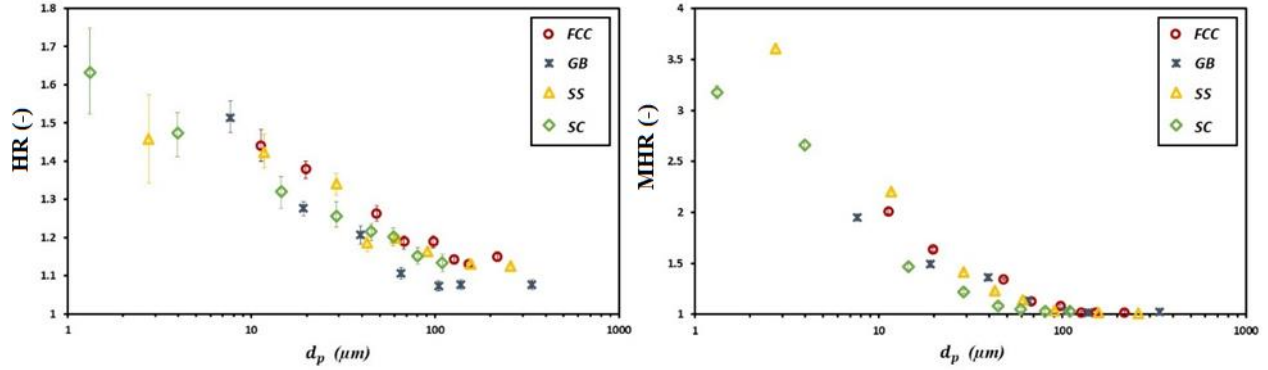


Figure 7.2. Variations of HR and MHR with particle diameter.

Based on the experimental results in Figure 7.2 and Figure 7.3, and depending on the Geldart behaviour of particles, the following scenarios can be considered for the behaviour of the bed voidage in different states and corresponding HR and MHR:

- Geldart groups B and D:** At ambient conditions, Geldart groups B and D behaviours correspond to particles greater than about $100 \mu m$ where the magnitude of IPFs is insignificant, i.e., $IPFs/W_p < \sim 0.1$ [14]. The insignificance of IPFs for these particles is reflected in the similarity of ε_0 and ε_w as the latter is an indication of the particles' packing structure in the virtual absence of IPFs. The unimportance of IPFs has several implications for the behaviour of these particles. First, they do not form agglomerates. Second, ε_0 and ε_{mf} are very close. The difference between ε_0 and ε_{mf} is caused by IPFs [7] and results in the appearance of an overshoot at U_{mf} in the bed pressure drop profile when gradually increasing the gas flow [7]. Accordingly, no or minimal pressure drop overshoot is observed for these particles [7]. Not only Figure 7.1 illustrates the similarity of ε_0 , ε_{mf} , and ε_w for particles with Geldart groups B and D, but also the independence these parameters to the particle size. This can be theoretically deduced from the conformal symmetry with respect to d_p in geometry of systems with identical particles. In other words, in the absence of IPFs, the packing structure of particulate systems with identical particles must be independent of the length scale. Experimental results showed that the values of ε_0 , ε_{mf} , and ε_w for the

Geldart groups B and D particles are functions of the particle's shape and vary in the range of about 0.4 to 0.55. Values as low as about 0.35 for ε_0 have also been reported in literature [21, 22]. In addition, the values of ε_0 and ε_{mf} were greater for particles with smaller shape factors as reported in literature [20]. While these parameters were less than about 0.45 For FCC and GB with shape factors greater than 0.9, they were greater than about 0.5 for SiO₂ and SiC particles. Figure 7.1 shows that ε_{tap} is always smaller than ε_0 , even for particles with Geldart groups B and D behaviours. The difference between ε_0 and ε_{tap} results in HR of up to about 1.15 for these particles. This implies that the deviation of HR from one is not entirely because of IPFs. MHR, on the other hand, is very close to one for particles in these classes of behaviour.

- **Geldart group A:** The magnitudes of IPFs and W_p are comparable for these particles ($\sim 0.1 < IPFs/W_p < \sim 10$ [14]) with typical diameters of $20 - 100 \mu m$ at ambient conditions. The smaller the particle diameter, the greater the magnitude of IPFs. The increasing magnitude of IPFs can also be deduced from the progressive deviation of ε_0 from ε_w with decreasing d_p . The increase in ε_0 and ε_{mf} with decreasing d_p for Geldart groups A and C must be attributed to the growth in the magnitude of IPFs as these parameters did not change with d_p for Geldart groups B and D behaviours. This conclusion is in accordance with the reported increase of the voidage of a particulate material under an enhanced level of IPFs, for example, at high temperatures [7, 23, 24] or due to polymer coating [25]. Another consequence of the presence of an appreciable magnitude of IPFs on Geldart group A behaviour is the deviation of ε_{mf} from ε_0 that causes the appearance of a pressure drop overshoot at minimum fluidization [7]. Such magnitudes of IPFs for Geldart group A particles can also lead to the formation of agglomerates with mean diameters as large as about three times greater than the diameter of their primary particles [3, 14]. The presence of IPFs in a bed of Geldart group A particles leads to the deviation of $U_{mf,exp}$ from its predicted value by conventional correlations as they are incapable of considering the effects of these forces [26].

The variations of ε_{tap} and ε_0 with d_p typically result in a larger HR for particles with a greater magnitude of IPFs. In compliance with the criteria suggested by Geldart [10], the results of this study showed that HR could be up to about 1.4 for group A particles. The values of MHR for these particles varied in the range of 1 to about 1.6.

- **Geldart group C:** The transition from Geldart group A to C corresponds to the dominance of IPFs ($IPFs/W_p > \sim 10$ [3, 14]) and the formation of larger agglomerates ($d_{agg}/d_p > \sim 3$ [3, 14]). It typically occurs when $\varepsilon_{mf} \approx 0.6 - 0.7$ [14]. Channeling is the distinct characteristic of Geldart group C behaviour and the reason for the similarity of the ε_0 and ε_{mf} for these particles [3]. By decreasing particle size in this class of Geldart behaviours, the magnitude of IPFs increases dramatically (see Figure 7.5). Under these circumstances, the formation of larger agglomerates with greater porosity leads to a substantial increase in the bed voidage. Accordingly, ε_0 and ε_{mf} can reach values as high as about 0.99 for particles with very small d_p like nanoparticles [27, 28].

Like the observed trend for ε_0 and ε_{mf} , tapped bulk density increases sharply with decreasing particle diameter in the Geldart group C region. The variation of ε_0 and ε_{tap} for these particles leads to HR greater than about 1.4. While reported values of HR are smaller than 2.2 for micron-size particles [10], MHR values up to about four were observed in our experiments. In addition, the uncertainty in the measurement of HR is considerably greater than that for MHR in the case of Geldart group C powders. This is due to the propagation of uncertainties in the measurement of both ε_{tap} and ε_0 into HR, while MHR is only affected by ε_{tap} with relatively smaller uncertainty compared to ε_0 .

The above remarks are summarized in Table 2.

Table 7.2. Typical properties of the bed voidage in different states based on the Geldart behaviour and corresponding agglomerate size and magnitude of IPFs.

Geldart groups B and D	Geldart group A	Geldart group C
$IPFs/W_p < \sim 0.1$	$0.1 < IPFs/W_p < \sim 10$	$IPFs/W_p > \sim 10$
$d_{agg,mf}/d_p \approx 1$	$1 < d_{agg,mf}/d_p < \sim 3$	$d_{agg,mf}/d_p > \sim 3$
• $\varepsilon_{mf} \approx \varepsilon_0 \approx \varepsilon_w > \varepsilon_{tap}$ • $\sim 0.35 < \varepsilon_{mf} \approx \varepsilon_0 \approx \varepsilon_w < \sim 0.55$ (Depending on the shape of the particles). • ε_{mf}, ε_0, ε_w, and ε_{tap} are independent of the particle size. • $1 < HR < \sim 1.15$. • $MHR \approx 1$.	• $\varepsilon_{mf} > \varepsilon_0 > \varepsilon_w > \varepsilon_{tap}$. • ε_w cannot be adequately measured for finer particles in this group. • ε_{mf}, ε_0, and ε_{tap} increase with decreasing particle size. • $\sim 1.1 < HR < \sim 1.4$. • $1 < MHR < \sim 1.6$.	• $\varepsilon_0 > \sim 0.6$. • $\varepsilon_{mf} \approx \varepsilon_0 > \varepsilon_{tap}$. • ε_{mf}, ε_0, and ε_{tap} increase more dramatically with decreasing particle size. • $\sim 1.4 < HR < \sim 2.2$ (For micron-size particles). • $MHR > 1.6$.

7.4.2 Performance of the MHR equation

7.4.2.1 Prediction of the MHR model versus data in literature

Figure 7.3 compares the magnitude of IPFs estimated by the MHR equation with the experimental data in literature [1, 3, 5, 7, 23-25, 29-38] and those from the current study. The predictions of the MHR equation were calculated by estimating MHR with Eq. 7.17 and ε_{i-agg} in the range of 0.29-0.55, as suggested by Eq. 7.27. To calculate the magnitude of IPFs, the value of D_a was initially considered 2.5, leading to the equivalence of the general and simplified MHR equations, i.e., Eqs. 7.14 and 7.16. The calculated magnitude of IPFs assuming $D_a = 2.6$ is also shown in Figure 7.3.

The predictions of the MHR model for the Geldart group A region are shown with dashed lines since this model was originally developed for Geldart group C powders.

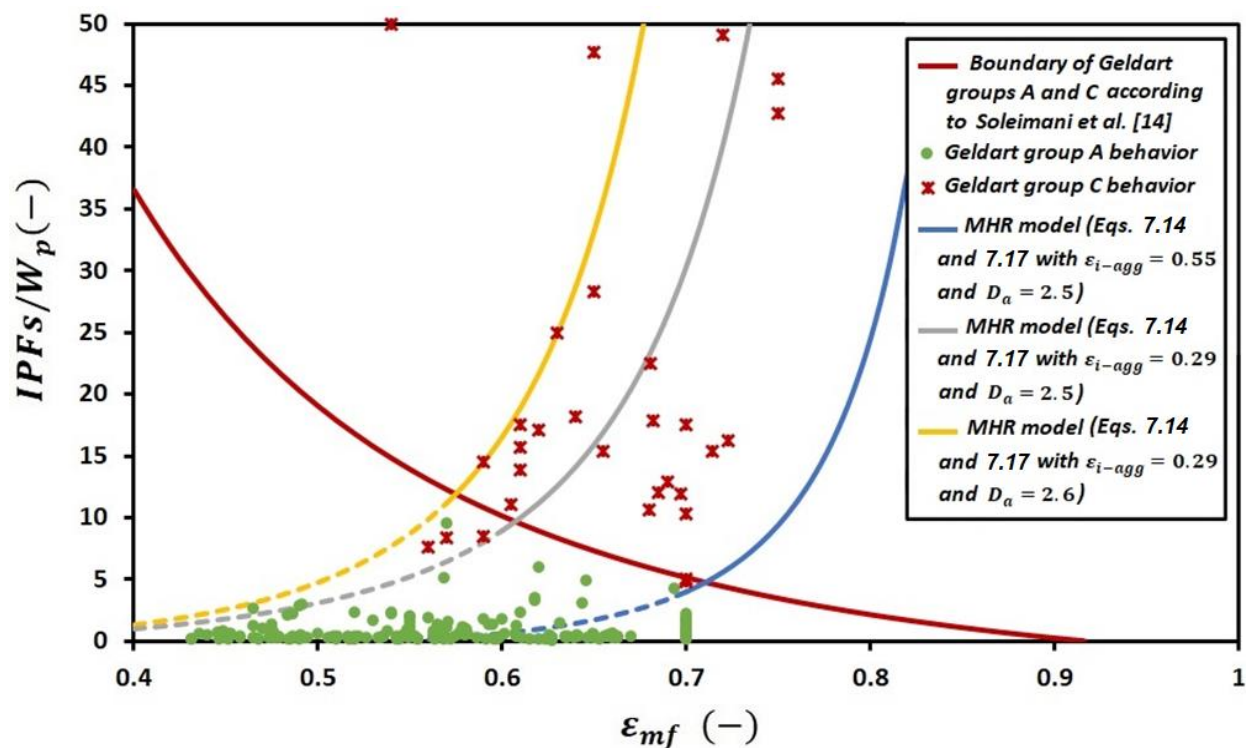


Figure 7.3. The predicted range of the magnitude of IPFs by MHR equation compared with experimental data in this work and literature.

As illustrated in Figure 7.3, the MHR model predicts that the magnitude of IPFs generally grows with bed voidage. This is consistent with experimental data of $IPFs/W_p$ – which were quantified by U_{mf} deviation equation – that are mostly within the predicted range by the MHR model. It also indicates the high sensitivity of the MHR equation to the fractal dimension of agglomerates and suggests that the value of D_a might be slightly greater than 2.5 in some cases.

Although the MHR model was developed based on the characteristics of Geldart group C powders, Figure 7.3 suggests that it can also generate practical results for particles with group A behaviour.

For these particles, however, the MHR model must be viewed as an interpolation between two valid states: Group C behaviour, where the MHR model was developed, and Groups B and D particles with negligible magnitudes of IPFs. Hence, applying the MHR equation to particles with Geldart Group A behaviour involves significant uncertainty.

7.4.2.2 The transition between Geldart groups A and C

We proposed the following equation for the magnitude of IPFs at the boundary between groups A and C in our recent study [14]:

$$\frac{IPFs}{W_p} = \frac{10 (1 - \varepsilon_{mf})}{\varepsilon_{mf}^2} - 1 \quad 7.28$$

The proposed criterion suggests that the magnitude of IPFs at the boundary between Geldart groups A and C varies with the bed voidage according to Eq. 7.28. It has no restriction on the value of ε_{mf} and corresponding magnitude of IPFs [14]. However, experimental results in Figure 7.3 show that the transition occurs when the bed voidage and magnitude of IPFs reach certain values. The MHR model can be combined with the above criterion to justify this observation. This model proposes a relationship between the magnitude of IPFs and ε_{mf} according to Eq. 7.16. Equating the left-hand sides of Eqs. 7.16 and 7.28 results in:

$$MHR^4 = \frac{10 (1 - \varepsilon_{mf})}{\varepsilon_{mf}^2} \quad 7.29$$

By replacing MHR from Eq. 7.17, we obtain:

$$\left(\frac{1 - \varepsilon_{i-agg}}{1 - \varepsilon_{mf}} \right)^4 = \frac{10 (1 - \varepsilon_{mf})}{\varepsilon_{mf}^2} \quad 7.30$$

Given that the value of ε_{i-agg} must be the range of about 0.29-0.55 for Geldart group C particles, Eq. 7.30 can be solved for ε_{mf} , leading to $\sim 0.61 < \varepsilon_{mf} < \sim 0.71$. Substituting these values in Eq.

7.16 results in the magnitude of IPFs in the range of $\sim 5 < IPFs/W_p < \sim 10$ which corresponds to $\sim 1.55 < MHR < \sim 1.8$ and $\sim 2.4 < d_{agg,mf}/d_p < \sim 3.2$. The calculated values of ε_{mf} and $IPFs/W_p$ agree with the experimental data for the transition between Geldart groups A and C shown in Figure 7.3. Please refer to Figure 7.2 and the corresponding discussion for the observed values of MHR for different Geldart groups. The estimated values of $d_{agg,mf}/d_p$ will be discussed in the next section.

7.4.2.3 Quantification of agglomerate size

The estimated ratios of $d_{agg,mf}/d_p$ by the corresponding MHR equation, Eq. 7.15, are presented in Figure 7.4. The results show that $d_{agg,mf}/d_p$ increases with decreasing particle diameter, especially for particles with Geldart group C behaviour, i.e., where $d_p < \sim 20 \mu m$. They also confirm the previously calculated $d_{agg,mf}/d_p$ corresponding to the boundary between Geldart groups A and C, i.e., $\sim 2.4 < d_{agg,mf}/d_p < \sim 3.2$ as particles with $d_{agg,mf}/d_p$ of about two still behave like Geldart group A (FCC-48, GB-19, SS-29, and SC-15), whereas $d_{agg,mf}/d_p$ is greater than about three for Geldart group C powders (FCC-20, FCC-11, GB-8, SS-12, SS-3, SC-4, and SC-1). These results also agree with experimentally measured agglomerate size for Geldart groups A and C powders with high-speed cameras [39, 40] and an optical fiber probe [3].

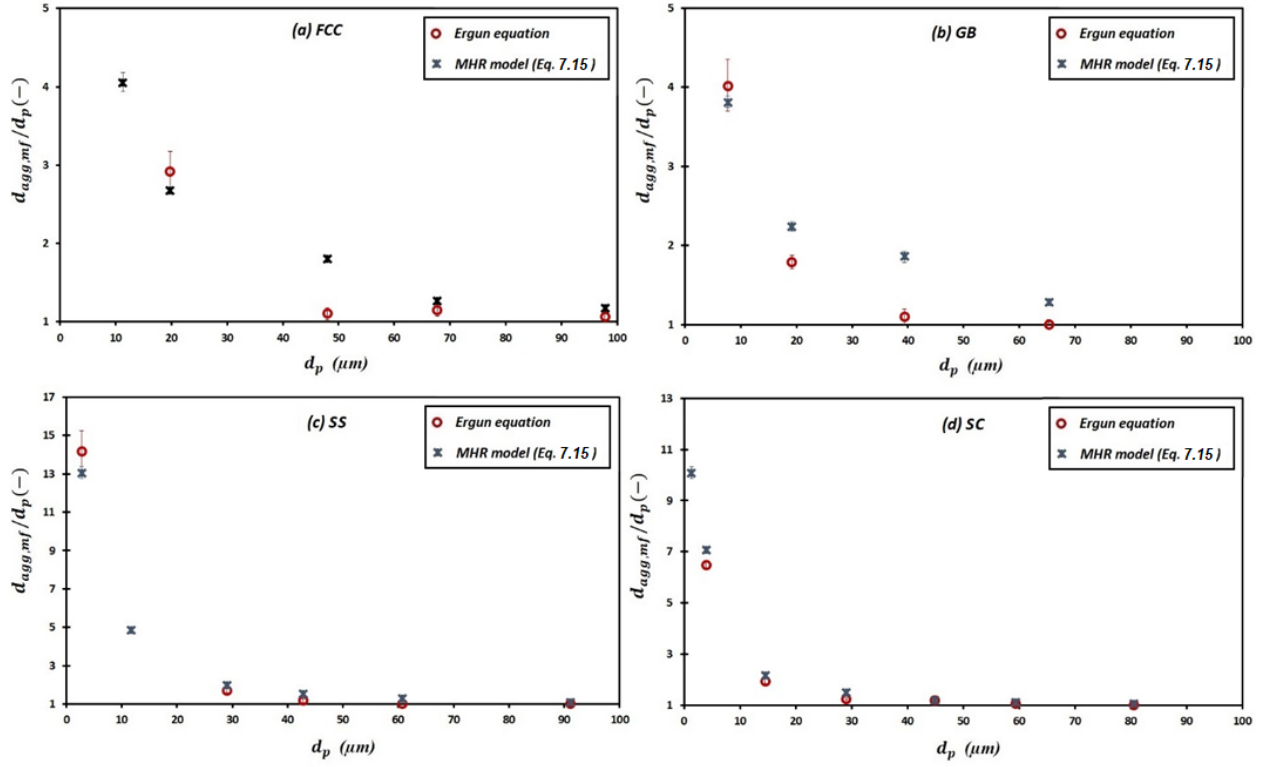


Figure 7.4. Variation of $d_{agg,mf}/d_p$ with particle diameter.

By conducting experimental measurements of $U_{mf,exp}$, along with the corresponding bed height, L_{mf} , pressure drop, ΔP_{mf} , and voidage, we estimated the effective diameters of the agglomerates under minimum fluidization conditions. This estimation was achieved by rearranging the viscous term of the Ergun equation as follows:

$$d_{agg,mf} = \sqrt{\frac{150 U_{mf,exp} \mu_g L_{mf} (1 - \varepsilon_{mf})^2}{\varepsilon_{mf}^3 \Delta P_{mf}}} \quad (Re_{p,mf} < 20) \quad 7.31$$

Eq. 7.31 is valid only when the pressure drop is dominated by the viscous effect, which corresponds to Particles' Reynolds number, $Re_{p,mf}$, smaller than 20 [20].

The results in Figure 7.4 indicate that the agglomerate sizes predicted by the MHR equation agree with the experimentally measured agglomerate sizes by the pressure drop data and the Ergun

equation. This agreement suggests that the MHR equation can effectively predict the agglomerate size. This estimation only requires the measurement of the tapped bulk voidage for Geldart group C powders and is considered the validation of the MHR equation for estimating agglomerate size.

7.4.2.4 Quantification of the magnitude of IPFs

The quantified magnitudes of IPFs by Eq. 7.16 are presented in Figure 7.5. Except for powders with extreme Geldart group C behaviour that could not be normally fluidized, the magnitudes of IPFs were also experimentally measured employing U_{mf} deviation approach with the following equation [3]:

$$\frac{IPFs}{W_p} = \frac{U_{mf,exp}}{U_{mf,pred}} - 1 \quad 7.32$$

where $U_{mf,pred}$ is the predicted value by the Carman-Kozeny equation [41] as a purely hydrodynamic correlation:

$$U_{mf,pred} = \frac{d_p^2}{C_{C-K}} \frac{\rho_p - \rho_g}{\mu_g} g\left(\frac{\varepsilon_0^3}{1 - \varepsilon_0}\right) \quad 7.33$$

where C_{C-K} is the constant of the Carman-Kozeny equation, usually about 180, and μ_g and ρ_g are the gas viscosity and density, respectively.

Figure 7.5 shows that IPFs grow significantly with decreasing d_p and become at least about an order of magnitude greater than W_p for Geldart group C powders. These results agree with the criterion for the transition between Geldart groups A and C as expressed by Eq. 7.28 and shown in Figure 7.3. Figure 7.5 also demonstrates a good agreement between the results of the MHR equation and U_{mf} deviation approach, validating the MHR equation for quantifying the magnitude of IPFs.

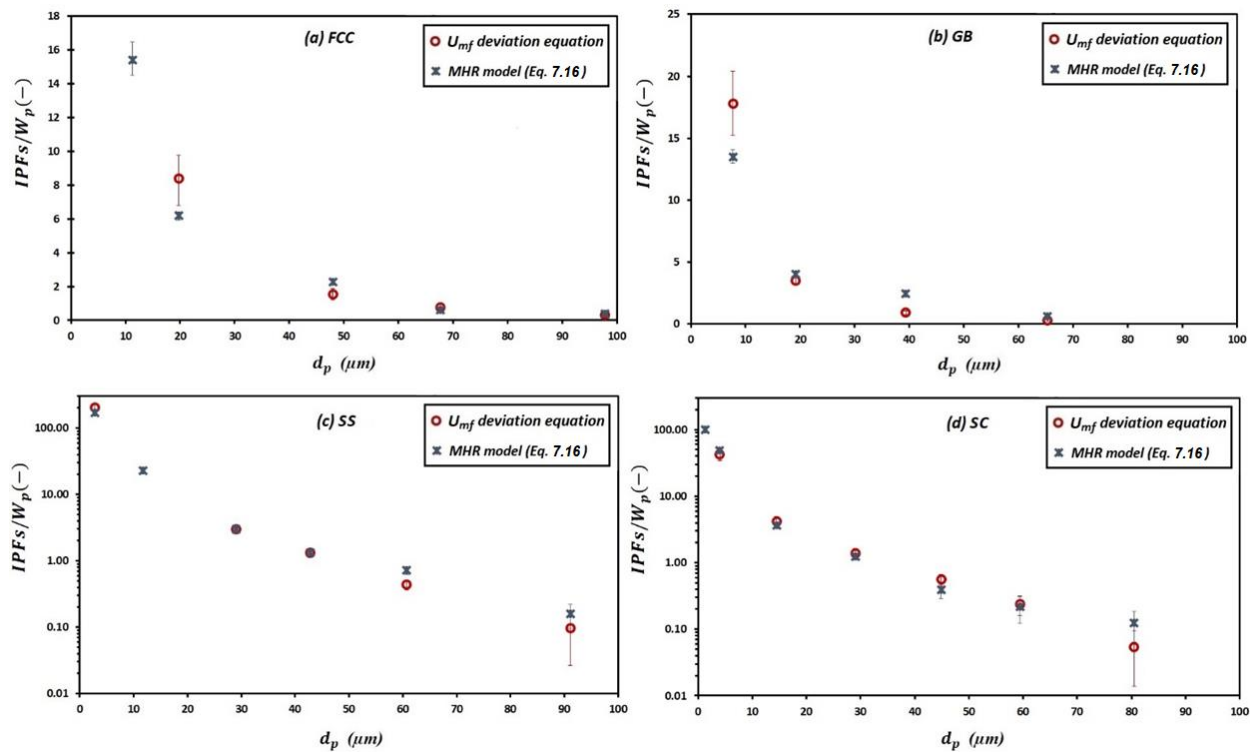


Figure 7.5. Variation of the magnitude of IPFs with particle diameter.

7.4.3 Uncertainties of MHR equation

Particle size distribution: The MHR model is not valid for powders with wide or bimodal size distributions since these conditions are inconsistent with the adopted assumptions to develop this model. A common example is adding ultra fine or nanoparticles to a micron size cohesive particulate material with Geldart group C behaviour to improve its fluidization characteristics. Under these circumstances, finer particles move in between coarser ones and act like a lubricant. Consequently, the bed voidage and macro scale cohesivity decreases and a more favorable fluidization behaviour is observed. The bed voidage under these conditions decreases despite the fact that the magnitude of IPFs is greater for finer particles.

Fractal dimension: The general form of the MHR equations, i.e., Eqs. 7.13 and 7.14, require knowledge of the fractal dimension of agglomerates. The dependence of the general form of MHR

equations to D_a is a strong point in favor of this model from a scientific point of view. However, one of the greatest uncertainties in the results of the MHR equations comes from their high sensitivity to the value of the fractal dimension. Although researchers have shown that the fractal dimension is nearly 2.5 for micron-size particles [12], even a small uncertainty in this value can lead to large uncertainties in agglomerate size and, particularly, the magnitude of IPFs. For instance, when MHR is equal to 1.75, i.e., for particles close to the boundary of Geldart groups A and C, adopting 2.4 and 2.6 instead of 2.5 for the fractal dimension in Eqs. 7.13 and 7.14 lead to estimating $d_{agg,mf}/d_p$ of 2.54 and 4.05, respectively. They also result in the quantification of $IPFs/W_p$ equal to 5.46 and 15.41, respectively.

Nanoparticles: One of the uncertainties in applying the general forms of the MHR equations for nanopowders is due to the wide varying fractal dimension of nanoparticle agglomerates. While D_a is practically 2.5 for micron-size particles [12], it can have values of about 1.75-2.2 for nanoparticle agglomerates [13]. Given the high sensitivity of the results of the general MHR equations to the value of D_a , its appropriate value must be known for a reliable application of the MHR equations.

Particles with Geldart group A behaviour: Some assumptions adopted for developing the MHR equations are only valid for particles with Geldart group C behaviour. For example, it was assumed that ε_{mf} and ε_0 are equal, leading to the simplified DHR equation, Eq. 7.3, which was subsequently employed to develop Eqs. 7.14 and 7.16. As alluded to earlier, applying the MHR equations for Geldart group A particles must be fundamentally considered as an interpolation of this equation between two valid states. They include Geldart group C powders, where MHR equations were developed, on the one hand, and Geldart group B and D particles with Negligible magnitude of IPFs, on the other. Nevertheless, the results of the MHR equations for Geldart group A particles in this study were consistent with the results of another experimental technique, i.e., the U_{mf} deviation approach.

Limitations of the DHR equation: The DHR equation served as the basis of the MHR equation for quantifying IPFs. It has limitations under the following circumstances: (i) when the bed voidage is above 0.8, where the constant of the Ergun equation can significantly change from its conventional value (=150) [42], and (ii) if particles' Reynolds number is greater than about 20,

where the inertia effect cannot be neglected [20], and (iii) for nanoparticles due to the gas screening effect [9], i.e., the tendency of the gas to predominantly flow through the intra-agglomerate rather than inter-agglomerate voids. Under these conditions, the DHR equation and, consequently, the MHR equations for estimating the magnitude of IPFs become invalid. However, the estimation of agglomerate size by the corresponding MHR equation may still be valid under these circumstances.

7.5 Conclusion

This chapter presents the development and validation of a model that relates bed voidage to agglomerate size and the magnitude of IPFs. The model introduces a modified Hausner ratio (MHR) as a key parameter. For Geldart group C powders, MHR becomes equal to the ratio of particle density and the tapped bulk density.

The MHR model led to two equations. The first equation shows that the ratio of agglomerate and particle size is proportional to the square of MHR. The second indicates that the ratio of IPFs and weight of a particle varies with the fourth power of MHR.

The MHR equations exhibited promising performance, particularly for Geldart group C powders where IPFs govern the fluidization behaviour. The results showed that the diameter of agglomerates is at least about three times greater than primary particles with Geldart group C behaviour. They also indicated that IPFs acting on these particles are at least about an order of magnitude greater than the weight of these particles. MHR equations were validated by comparing their results with the agglomerate size estimated by pressure drop data and the Ergun equation and quantified IPFs by the U_{mf} deviation approach.

The presented model can be employed to improve the prediction of hydrodynamic correlations and simulations of gas-solid systems. The MHR equations can aid in adequately predicting hydrodynamic parameters by considering the effects of IPFs. It also offers a straightforward procedure for tracking the size of agglomerates as a crucial parameter responsible for bed defluidization under a significant level of IPFs. Furthermore, the experimentally measured IPFs by

the MHR equations can be employed in the discrete element method (DEM) to enhance the reliability of simulating gas-solid system behaviour.

The proposed equations have limitations when applied to nanoparticles and powders with wide size distributions, requiring further development for more accurate predictions in such cases.

Acknowledgements

The authors express their sincere gratitude for the financial support from OCP Group and Mitacs through the Mitacs Accelerate program.

7.6 Nomenclature

Acronyms

DEM	discrete element method
DHR	dynamic Hausner ratio
FCC	fluid cracking catalyst
GB	glass beads
HR	Hausner ratio
IAFs	inter-agglomerate forces
IPFs	interparticle forces
MHR	modified Hausner ratio
SC	Silicon carbide
SS	silica sand

Symbols

C_{C-K}	constant in the Carman–Kozeny equation (-)
D_a	fractal dimension of agglomerates (-)
$d_{agg,mf}$	effective agglomerate diameter at minimum fluidization conditions (m)
d_p	particle diameter (m)
$IPFs$	interparticle forces acting on each particle (N)
L_{mf}	bed height at minimum fluidization conditions (m)
$N_{p,agg}$	number of particles in an agglomerate (-)
$Re_{p,mf}$	particle Reynolds number at minimum fluidization conditions (-)
U_{mf}	minimum fluidization velocity (m/s)
$U_{mf,exp}$	experimental minimum fluidization velocity (m/s)
$U_{mf,pred}$	minimum fluidization velocity predicted by the Carman-Kozeny equation (m/s)
$V_{agg,mf}$	volume of an agglomerate at minimum fluidization (m ³)
V_p	volume of a particle (m ³)
W_{agg}	weight of a single agglomerate (N)
W_p	weight of a single particle (N)

Greek letters

ΔP_{mf}	bed pressure drop at minimum fluidization conditions (Pa)
ε_0	loose bulk voidage (-)

ε_{agg}	voidage of an agglomerate (-)
$\varepsilon_{agg,mf}$	voidage of an agglomerate at minimum fluidization (-)
ε_{i-agg}	intra-agglomerate voidage (-)
ε_{mf}	minimum fluidization voidage (-)
ε_{tap}	tapped bulk voidage (-)
ε_w	water phase fraction in water-solid system (-)
μ_g	gas viscosity (Pa. s)
ρ_0	loose bulk density (kg/m ³)
ρ_g	gas density (kg/m ³)
ρ_p	particle density (kg/m ³)
ρ_{tap}	tapped bulk density (kg/m ³)

CHAPTER 8 GENERAL DISCUSSION

Quantifying IPFs in a gas-solid fluidized bed and determining the powders' Geldart group behaviour based on the relative magnitude of these forces compared to HDFs were set as the main objective of this work. The first part of this study was dedicated to developing in-situ approaches to reliably estimate effective IPFs in a gas-solid fluidized bed. In the second section, the findings in the first section were employed to include the effect of IPFs in the Geldart classification. The results in the second section provided a basis for an additional approach for quantifying IPFs.

U_{mf} is arguably the most important fluidization hydrodynamic parameter and is used in many correlations for the estimation of other hydrodynamic parameters, usually in the form of excess gas velocity, i.e., $U_g - U_{mf}$. It is the superficial gas velocity where the particles start to move relative to each other and, thus, particulate material begins to show fluid-like behaviour. Accordingly, the weight of the bed is required to be overcome by the HDFs generated by the gas flow at U_{mf} . In the case of cohesive particles, the HDFs at U_{mf} must overcome IPFs in addition to the weight of the particles. This concept was employed in the first section of this study to develop U_{mf} deviation and DHR approaches for in-situ quantification of IPFs.

In the U_{mf} deviation approach, the difference between the experimental and theoretical values of minimum fluidization velocity is converted into the gas pressure drop required to overcome IPFs and, ultimately, to the IPFs acting on each particle. In its generalized form, the U_{mf} deviation approach turned into a simple equation relating the ratio of IPFs and the weight of a particle ($IPFs/W_p$) to the ratio of experimental and theoretical minimum fluidization velocities ($U_{mf,exp}/U_{mf,pred}$). In the DHR approach, the magnitude of IPFs is directly calculated by the overshoot pressure drop that appears at U_{mf} for cohesive particles. In individual particle fluidization, the DHR equation is a relationship between $IPFs/W_p$ and the DHR, i.e., the ratio of $\varepsilon_{mf}/\varepsilon_0$. The relationship with the ratio of agglomerate and particle size, $d_{agg,mf}/d_p$ was integrated into the generalized form of the DHR equation to extend its applicability to agglomerating

fluidization. In the generalized DHR equation $\varepsilon_{mf}/\varepsilon_0$ signifies IPFs that are overcome during fluidization while $d_{agg,mf}/d_p$ represents IPFs that continue being exerted on particles after fluidization.

In addition to the above two approaches, a model was presented to correlate the bed voidage with $d_{agg,mf}/d_p$ and fractal dimension of agglomerates. A significant contribution is made by introducing a modified Hausner ratio (MHR) as a crucial parameter in this model, specifically for powders with Geldart group C behaviour, where the MHR is defined as the ratio of particle and tapped bulk densities. This model resulted in the development of two equations for quantification of $d_{agg,mf}/d_p$ and $IPFs/W_p$. These equations indicate that $d_{agg,mf}/d_p$ is directly proportional to the square of the MHR, while $IPFs/W_p$ is a function of the fourth power of the MHR.

The results showed that, in comparison with W_p , IPFs are negligible for particles with Geldart group B behaviour, while they are in the same order of magnitude if particles exhibit Geldart group A behaviour. In gas-solid fluidized beds with Geldart group C behaviour, IPFs acting on particles are at least about an order of magnitude greater than their weight and can be as high as three to four orders of magnitude greater than W_p . The variations of $\varepsilon_{mf}/\varepsilon_0$, $d_{agg,mf}/d_p$ and $IPFs/W_p$ with temperature for Geldart group A behaviour and particle diameter for Geldart groups A and C behaviours were investigated. The magnitude of IPFs slightly diminished up to about 100°C, likely due to the decrease of the electrostatic forces. Above this temperature, IPFs increased significantly. Particles essentially show individual fluidization behaviour, and the magnitude of IPFs was mainly reflected in $\varepsilon_{mf}/\varepsilon_0$ if IPFs are smaller than W_p . By increasing IPFs, larger agglomerates form, and the ratio of $d_{agg,mf}/d_p$ contributes to the magnitude of IPFs. It was shown that ε_{mf} and ε_0 increase with IPFs. For fine powders where IPFs dominate over W_p , ε_{mf} and ε_0 are principally controlled by the magnitude of IPFs rather than the physical characteristics of the particles. If van der Waals forces dominate the magnitude of IPFs, applying the Hamaker model with the particle's radius replaced by its asperity size produced comparable results with the proposed equations.

The strategy adopted in this study to quantify effective IPFs acting on particles in a gas-solid fluidized bed employs fluidization parameters at minimum fluidization conditions. The ultimate

results of this strategy, i.e., the generalized U_{mf} deviation, DHR, and MHR equations showed promising performances in adequately estimating the magnitude of IPFs and were also verified and qualitatively validated by different principles. The primary advantage of this strategy is to avoid inconsistent or unrealistic values of IPFs produced by ex-situ approaches, which may not be applicable to a gas-solid fluidized bed. The simplicity of the measurement of minimum fluidization velocity and tapped bulk voidage make the U_{mf} deviation and MHR equations the primary choices for quantifying IPFs for Geldart groups A and C, respectively. The DHR equation reveals the underlying effects of IPFs on the bed voidage and agglomerate size.

The U_{mf} deviation and DHR equations are not predictive, i.e., a fluidization test is required for the measurement of U_{mf} and the corresponding bed voidage and agglomerate size for quantifying IPFs to be subsequently employed to estimate other hydrodynamic parameters, including the Geldart behaviour. Although methodologies were suggested in this study to predict the magnitude of IPFs for different particles knowing the fluidization parameters for one, more work is needed to fully develop these approaches, especially when sintering or liquid bridge forces are involved. Moreover, the U_{mf} deviation and DHR equations cannot be applied to many cohesive particles with Geldart group C behaviour since they are not normally fluidizable. However, MHR equations provide a straightforward method for these circumstances. Other shortcomings of the proposed equations include quantifying IPFs for nanoparticles, if the bed voidage exceeds 0.8, and beyond the viscous flow regime. IPFs cannot be neglected in most of these circumstances. Hence, more research is required to address these limitations.

As their first application for the prediction of bed hydrodynamics, the Generalize U_{mf} deviation and DHR equations were employed to develop a fundamental model to characterize the boundary between Geldart groups A and C behaviours. The outcome of the model was a relationship between $IPFs/W_p$ and ε_{mf} at the boundary between Geldart groups A and C. According to experimental data, ε_{mf} in the range of 0.6 to 0.7, which corresponds to IPFs about an order of magnitude larger than W_p and agglomerates about three times greater than the particle size are typical characteristics of the transition point between these two Geldart groups. The particle diameter at which the transition between Geldart groups A and C happens was demonstrated to be proportional to the

cube root of IPFs. Despite the successful discriminating of Geldart groups A and C behaviours based on the magnitude of IPFs, further development is needed for distinguishing agglomerate particulate fluidization and agglomerate bubbling fluidization as common behaviours in nanoparticle fluidization.

The global need for more efficient processes is leading to a situation where the operation of gas-solid fluidized beds under an enhanced level of IPFs is more frequently experienced. These conditions include the utilization of finer particles with greater specific surface area and high-temperature processing of low-quality feedstocks like waste. The proposed equations for the quantification of IPFs can help in the development of reliable hydrodynamic correlations that take into account the effect of IPFs in addition to HDFs. These correlations are essential for the proper design and scale-up of gas-solid fluidized beds. In addition, the accuracy of CFD-DEM models in the simulation of these units can be improved with an accurate estimation of IPFs by the proposed equations.

CHAPTER 9 CONCLUSION AND RECOMMENDATIONS

9.1 Concluding remarks

Three approaches were developed in this study to quantify the magnitude of IPFs in a gas-solid fluidized bed. The U_{mf} deviation approach is established based on the idea that IPFs cause the experimental minimum fluidization velocity to deviate from its theoretical value. This approach led to a linear relationship between the magnitude of IPFs and the ratio of experimental and theoretical minimum fluidization velocities. The overshoot pressure drop at U_{mf} in the increasing velocity pass, as the additional gas pressure drop required to overcome IPFs, was the origin of the second approach. The resulted equation from the proposed model for this overshoot correlates IPFs with the ratio of $\varepsilon_{mf}/\varepsilon_0$ which was called the dynamic Hausner ratio (DHR). The original DHR equation does not consider the IPFs that are not overcome during fluidization, i.e., those between particles within agglomerates. Hence, it underestimates the magnitude of IPFs under agglomerating fluidization conditions. The generalized version of the DHR equation was subsequently developed based on the equivalence of the U_{mf} deviation and DHR equations. It correlates the magnitude of IPFs with the ratio of agglomerate and particle sizes in addition to the DHR. The third approach was founded on the observed relationship between the magnitude of IPFs and bed voidage. The bed voidage increases with the formation of larger agglomerates due to greater IPFs. A model was proposed for this relationship, leading to two equations for quantifying the magnitude of IPFs and agglomerate size with a parameter called modified Hausner ratio (MHR). MHR is nearly one for Geldart groups B and D with insignificant magnitudes of IPFs and becomes equal to the ratio of particle density to the density of the tapped bulk for Geldart group C powders. The experimental results showed that $IPFs/W_p < \sim 0.1$ for Geldart groups B and D, $\sim 0.1 < IPFs/W_p < \sim 10$ for Geldart group A, and $IPFs/W_p > \sim 10$ for group C.

A fundamental model for the prediction of the boundary between Geldart groups A and C behaviours was presented based on the magnitude of IPFs quantified by the proposed U_{mf}

deviation and DHR equations. The model suggests that the particle diameter at this boundary is proportional to the cube root of IPFs. It predicts that the magnitude of IPFs and the ratio of agglomerate and particle diameters corresponding to the boundary vary with ε_{mf} . The model showed excellent agreement with experimental data, which indicated that the transition between Geldart groups A and C behaviours are usually observed at $0.6 < \varepsilon_{mf} < 0.7$. At this range of ε_{mf} , IPFs are about an order of magnitude greater than W_p and agglomerates are about three times larger than the particles.

A limitation analysis of the proposed models suggests that the U_{mf} deviation, DHR, and MHR equations are invalid under the inertial flow regime and may lead to inadequate estimation of IPFs for nanoparticles. In addition, the proposed boundary between Geldart groups A and C must be further developed for distinguishing between agglomerating particulate and agglomerating bubbling fluidization behaviours experienced in nanoparticle fluidization and the behaviour of micron-size Geldart group C powders.

9.2 Original contributions

The present study provides valuable insight and crucial knowledge regarding the effect of IPFs on the hydrodynamic parameters of gas-solid fluidized beds at minimum fluidization conditions. By proposing appropriate models for the variation of these parameters with IPFs, we derived three novel equations to quantify the magnitude of these forces and an equation to estimate the size of agglomerates. A new perspective on the hydrodynamics of gas-solid fluidized beds is initiated by the outcomes of this study, i.e., it became possible to quantitatively consider the effect of IPFs on the bed hydrodynamics thanks to the proposed equations as the main contribution of the present study. Apart from quantifying the magnitude of IPFs, the first achievement obtained with the help of the proposed equations is the prediction of the transition between Geldart groups A and C behaviours, which had been a subject of controversy for about 50 years. The specific novelties of this research study are summarized as follows:

- An understanding of the variation of minimum fluidization velocity, corresponding bed voidage, and formation of agglomerates with IPFs, which led to phenomenological models for the quantification of IPFs.
- The generalized U_{mf} deviation equation which correlates the ratio of IPFs and the weight of a particle with the ratio of experimental and predicted minimum fluidization velocities.
- The generalized Dynamic Hausner ratio (DHR) equation, which correlates the ratio of IPFs and the weight of a particle with the ratio of minimum fluidization and loose bulk voidages and the ratio of agglomerate and particle diameters.
- The MHR equation for the estimation of the ratio of agglomerate and particle diameters with bed voidage.
- The MHR equation for quantification of the magnitude of IPFs with bed voidage.
- A fundamental model and equation correlating the boundary between Geldart groups A and C to the magnitude of IPFs.
- A novel equation to predict agglomerate size at the boundary between Geldart groups A and C.
- A novel experimental procedure for measuring agglomerate size by optical fiber probe.
- Experimental results on the magnitude of IPFs and its variation with temperature. The magnitudes of IPFs at ambient conditions were quantified for various particulate materials with Geldart groups B, A, and C behaviours. The data for the variation of IPFs with temperature were generated for particles with Geldart group A behaviour.

9.3 Recommendations for future studies

In this research, in-situ quantification of IPFs in a gas-solid fluidized bed was explored based on the effect of these forces on the hydrodynamic parameters at minimum fluidization conditions. In addition to the knowledge obtained here, the following research directions are suggested:

- **Correlations between the magnitude of IPFs quantified by different approaches:** The magnitude of IPFs acting on particles depends on many parameters, including the separation distance, whose average value can be considered a function of the bed voidage. This cause the quantified IPFs by different approaches to vary significantly as they are applied on particles under different conditions (tapped, aerated, under different normal stress, etc.). The recommended development makes it possible to adequately estimate IPFs under various conditions by applying one measurement technique.
- **Extension of the U_{mf} deviation and DHR equations for quantification of IPFs for nanoparticles:** Quantification of IPFs for nanoparticles is one of the most important limitations of the proposed equations. A basis for extending the proposed equations for quantifying IPFs in nanoparticle fluidization was presented in Section 5.5.5.
- **Discriminating agglomerating particulate and agglomerating bubbling fluidization from the behaviour of micron-size Geldart group C particles:** These two fluidization behaviours are typically observed in nanoparticle fluidization. Distinguishing ABF and APF from Geldart group C behaviour must be addressed, considering the significance of nanoparticles and their applications.
- **Develop models and correlations for the prediction of the bed hydrodynamic parameters considering the effects of both IPFs and HDFs:** Developing such models/correlations was, in fact, one of the initial motivations of this study. They are critically important for the appropriate design of gas-solid fluidized beds at elevated operating conditions, where both IPFs and gas properties - hence, HDFs - differ from ambient conditions.

- **Investigation and optimization of the effects of IPFs on the performance of a catalytic gas-solid fluidized bed reactor:** The DHR equation suggests that IPFs have two opposing effects on the performance of gas-solid fluidized beds: an increase in the bed voidage and formation of agglomerates. The former enhances the reactor performance by increasing the interstitial gas velocity, while the latter reduces the gas-solid contact and, thus, the overall reaction rate. Finding the optimum level of IPFs where the reaction rate is maximized can enhance the performance and improve the design of a gas-solid fluidized bed.
- **Investigation of the effects of thermally induced IPFs caused by the temperature difference between gas and solid on the performance of microwave-heated fluidized bed reactors:** One of the crucial advantages of microwave heating is providing a temperature gradient between solid and gas phases with higher solid temperature. These conditions result in a faster reaction rate on the surface of the solid while hindering unfavorable side reactions in the gas phase. At the same time, higher solid temperatures can also contribute to a greater magnitude of IPFs, leading to the formation of agglomerates that limit gas-solid contact and might eventually defluidize the bed. Research on this topic can assist in finding an optimum temperature difference between gas and solid phases to enhance the reaction rate without the danger of severe agglomeration and defluidization.

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