



# Prediction of the porosity of multi-component mixtures of cohesive and non-cohesive particles

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## ARTICLE INFO

### Article history:

Received 23 April 2011

Received in revised form

13 June 2011

Accepted 17 June 2011

Available online 25 June 2011

### Keywords:

Packing of particles

Porosity

Particle mixtures

Cohesive particles

Mathematical modeling

Packing similarity

## ABSTRACT

This paper presents an experimental and theoretical study of the packing of mixtures of cohesive (fine) and non-cohesive (coarse) particles. The experimental results, produced by means of a standard funnel packing method with glass beads as experimental materials, are first used to depict the similarity between the packings of fine and coarse particles. On this basis, the so-called linear packing model is extended to estimate the porosity of mixtures of fine and coarse particles with a wide size range. Its interaction functions and equivalent packing size are determined empirically. The applicability of the resultant model is demonstrated by the good agreement between the predicted and measured results for typical packing systems, including particles with the Gaudin–Schuhmann distribution or with a mixture size distribution. Finally, the packing of particles with a lognormal distribution, involving both cohesive and non-cohesive particles, is investigated in detail. This example also demonstrates how the proposed model can be used to solve a packing problem.

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## 1. Introduction

Particle packing, as a core research area in particle/powder technology, is of prime importance in many industries, material and chemical industries in particular (German, 1989). For example, densification of a powder mass is important in the shaping of solids in ceramics, powder metallurgy or composite synthesis; proper description of particle packings at either microscopic or macroscopic level is fundamental to many mineral or chemical processes such as solid–liquid separation (sedimentation, thickening and filtration), raw material handling (stockpiling, blending, drying, storage and transportation) and pyrometallurgical processes (agglomeration, coke making, ore sintering, blast furnace ironmaking and new ironmaking processes). It is very useful to understand the underlying fundamentals and on this basis, to develop a method for predicting the packing properties of particulate materials for property and/or process control.

Previous studies indicate that there are many factors affecting particle packing (German, 1989). However, as far as particle characteristics are concerned, a situation often encountered in practice, there are three main factors: (dimensionless) particle size distribution, particle shape and absolute particle size. These factors may lead to various complex packing systems from mono- to multi-sized, spherical to non-spherical, and/or from coarse

(non-cohesive) to fine (cohesive) particle packing. Understanding and modeling the relationship between porosity, the most commonly used packing property, and these particle characteristics has been a subject of research for years, progressing from the simple spherical particle packing to the complicated cohesive and non-spherical particle packing, as reviewed elsewhere (Yu and Zou, 1998; Latham et al., 2002). In general, in the 1980s research efforts are mainly made in modeling this relationship for non-cohesive or coarse spherical particles (Dodds, 1980; Ouchiya and Tanaka, 1986; Stovall et al., 1986; Yu and Standish, 1988, 1991). Such modeling efforts are then extended to the packing of non-spherical particles in the 1990s (Yu et al., 1993, 1996; Zou and Yu, 1996; Mayadunne et al., 1996; Sanchez et al., 1998; Finkers and Hoffmann, 1998; Liu and Ha, 2002; Dias et al., 2004). Modeling the packing of cohesive fine particles started in the late 1990s (see, for example, Yu et al., 1997), but to date, it is still an open and challenging research area.

When particle size is less than about 100  $\mu\text{m}$ , inter-particle force, which is the collective outcome of weak forces such as the van der Waals and electrostatic forces, becomes more important than the gravity force (Visser, 1989; Israelachvili, 1991). Consequently, fine particles are cohesive and their packing behavior is different from that of coarse particles. This difference may be highlighted by the variation of initial porosity (defined as the porosity of mono-sized particles) with particle size and the formation of aggregates or agglomerates (Milewski, 1987; Mizuno et al., 1991; Santomaso et al., 2003; Biesheuvel, 2000; Mallol et al., 2008; Liu et al., 2011). For non-cohesive spheres,

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it has been well established that their initial porosity is 0.36 for random dense packing and 0.4 for random loose packing (Scott, 1960). On the other hand, nanoparticles (sometimes referred to as ultra-fine particles) can give porosity as high as 0.99 (Mizuno et al., 1991). For mono-sized particles, the relationship between porosity and particle size has been established (Yu et al., 1997, 2003). Moreover, in recent years, simulation studies have been made to understand the underlying mechanisms in terms of inter-particle forces (Yen and Chaki, 1992; Yang et al., 2000, 2003; Dong et al., 2006). For mixtures involving cohesive or fine particles, to date, only limited efforts have been made in the formulation of predictive models or equations (Hoffmann and Finkers, 1995; Yu et al., 1997; Gan, 2002; Gane et al., 2006). The resulting equations are often empirical, with limited applicability. For example, the equation proposed by Hoffmann and Finkers (1995) is only applicable to pharmaceutical powders with a narrow lognormal size distribution. To overcome this limitation, Yu et al. (1997) made an attempt to establish a model framework to describe the packing of fine particles, which involves the use of two important concepts: (i) the initial porosity to take into account the packing of mono-sized particles; (ii) the equivalent packing size ratio as a measure of the particle–particle interaction in forming a packing. The use of these concepts has been verified for alumina powder of size range 0.1–60  $\mu\text{m}$ . But the model has a limited applicability. To date, for particles with a wide size range involving both cohesive (fine) and non-cohesive (non-cohesive) particles, no mathematical model can generally and reliably predict their porosity.

The purpose of this paper is to present an attempt to overcome this problem. This will be done by developing a method to link together the previous models for the packing of coarse or non-cohesive particles (Yu and Standish, 1991; Yu et al., 1996) or fine or cohesive particles (Yu et al., 1997). The validity of the new model will be examined by comparing the calculated and measured results for various particle systems.

## 2. Mathematical modeling

### 2.1. Model framework

The system considered is assumed to be composed of  $n$  components of uniform particle density, with each characterized by its initial porosity  $\varepsilon_i$  and nominal diameter  $d_i$  ( $i=1, 2, \dots, n$ ;  $d_1 \geq d_2 \geq \dots \geq d_n$ ). The so-called nominal size here can be any measurable particle size, ideally equivalent volume diameter. The initial porosity is the porosity of a component, as mentioned above. The porosity of this system  $\varepsilon$  should be a function of  $\varepsilon_i$ ,  $d_i$  and volume fraction  $X_i$ . According to the modified linear packing model, the specific volume of the mixture  $V$  ( $=1/(1-\varepsilon)$ ) can be determined by the following equation (Yu and Standish, 1991; Yu et al., 1996):

$$V = \text{Max}\{V_1^T, V_2^T, \dots, V_n^T\} \quad (1a)$$

where

$$V_i^T = \sum_{j=1}^{i-1} [V_j - (V_j - 1)g(d_i, d_j)]X_j + V_i X_i + \sum_{j=i+1}^n V_j [1 - f(d_i, d_j)]X_j \quad (1b)$$

where  $V_i$  are the initial specific volumes, equal to  $1/(1-\varepsilon_i)$  by definition. Eq. (1a) means that  $V$  equals the maximum of  $V_i^T$  ( $i=1, 2, \dots, n$ ).  $f(d_i, d_j)$  and  $g(d_i, d_j)$  are referred to as the interaction functions between components  $i$  and  $j$ , and depend on their sizes  $d_i$  and  $d_j$ . Although the system considered does not necessarily consist of spherical particles, to avoid possible confusion, this work will only consider the packing of spherical particles.

The use of Eq. (1) requires information about  $\varepsilon_i$ ,  $f(d_i, d_j)$  and  $g(d_i, d_j)$  *a priori*. It has been established that initial porosity is constant for coarse spherical particles, generally taking the value of 0.4 for loose random packing or 0.36 for dense random packing (Scott, 1960). However, for fine particles, their initial porosity increases with the decrease in particle size, and depends on the inter-particle force relative to the gravity force on a particle (Yu et al., 2003):

$$\varepsilon_i = 0.36 + 0.64 \exp \left[ -0.178 \left( \frac{\rho_p}{A} \right)^{0.129} d_i^{0.457} \right] \quad (2a)$$

for dense random packing, and

$$\varepsilon_i = 0.40 + 0.60 \exp \left[ -0.106 \left( \frac{\rho_p}{A} \right)^{0.156} d_i^{0.552} \right] \quad (2b)$$

for random loose packing, where  $\rho_p$  is particle density and  $A$  is the so-called Hamaker constant that is a material property used in the calculation of the van der Waals force. For fused quartz,  $A$  is equal to  $6.5 \times 10^{-20}$  J, which may be used for glass beads as an approximation (Israelachvili, 1991; Yu et al., 2003). The validity of such a formulation has been qualitatively verified in the simulation studies of fine or wet particles (Yang et al., 2000, 2003), where the cohesive force such as the van der Waals attraction or capillary force between particles can be directly quantified. Eq. (2) can generally be used to estimate  $\varepsilon_i$ . On the other hand, for a given packing system,  $\varepsilon_i$  can be measured readily and can be related to particle size  $d_i$ . To be more quantitatively comparable with the measured results, this approach is used in the present work.

Two methods can be used to determine the interaction functions  $f(d_i, d_j)$  and  $g(d_i, d_j)$ . By Method 1, the two functions are obtained by directly fitting Eq. (1) to the measured results of binary mixtures of components  $i$  and  $j$ . This may involve the experimental measurements for many binary packing systems in order to fully establish the dependence of the interaction functions on  $d_i$  and  $d_j$ . By Method 2, it is assumed that the two functions are only dependent on the small-to-large size ratio  $r_{ij}$  of the two components, as done for coarse spherical particles (Yu and Standish, 1991). For fine spherical particles, if their equivalent packing sizes  $d_{pi}$  are used in calculating  $r_{ij}$ , then the two functions are, respectively, given by (Yu et al., 1997)

$$f(r_{ij}) = (1 - r_{ij})^{3.33} + 2.81 r_{ij} (1 - r_{ij})^{2.77} \quad (3)$$

and

$$g(r_{ij}) = (1 - r_{ij})^{1.97} + 0.36 r_{ij} (1 - r_{ij})^{3.67} \quad (4)$$

The so-called equivalent packing diameter  $d_{pi}$ , like other equivalent spherical diameters, should be determined by relating the size-dependent packing property to the diameter of a sphere, as used for non-spherical particles (Zou and Yu, 1996). For coarse spherical particles,  $d_i = d_{pi}$ . But this relationship does not hold for fine particles. In general,  $r_{ij}$  ( $=d_{pj}/d_{pi}$ ;  $i < j$ ) should therefore be dependent on  $d_i$  and  $d_j$  or  $d_i$  and  $R_{ij}$  ( $=d_j/d_i$ ). The value of  $r_{ij}$  can again be obtained by fitting Eqs. (1)–(4) to the measured packing results of binary mixtures of components  $i$  and  $j$ . The two methods provide similar results. For convenience, only Method 2 is used in the present analysis.

### 2.2. Supportive experimental tests

Commercial glass beads were used in this work. Their density is around 2.5 g/cm<sup>3</sup>. In order to obtain glass beads with narrow particle size distributions, the beads were first classified by wet sieving, with beads less than 38  $\mu\text{m}$  in size further separated by a hydrocyclone apparatus. Twelve sized glass beads were obtained,

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