The Packing Density of Binary Powder Mixtures

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Abstract

The maximum particle packing density in a binary powder system can be predicted by the Furnas model. The packing density is lower when the size ratio of coarse particles to fine particles decreases, and is also governed by the volume fraction of coarse or fine particles. Here an empirical equation is derived which describes the packing density as a function of the initial packing efficiencies of particles, the particle size ratio, and the volume fraction variations of the system.

Introduction

The importance of particle packing in ceramic processing is generally appreciated because of its influence on the unfired mechanical properties, the shrinkage and density on sintering, and the microstructure and properties of the final ceramic product. A high maximum packing density* is directly dependent upon the particle size distribution. Many studies have demonstrated the importance of the particle size distribution to obtain dense packing.¹⁻¹⁴ For a two component mixture of coarse and fine particles, the ideal packing density is predicted by the Furnas model^{1,3,4} (and sometimes, the Westman and Hugill model²), (Fig. 1). The specific packing volume is defined as the reciprocal of the packing efficiency. According to the Furnas model, the theoretical maximum packing efficiency, PE_{max} , of a mixture of coarse and fine particles is:¹⁴

$$PE_{\text{max}} = PE_{\text{c}} + (1 - PE_{\text{c}})PE_{\text{f}} \tag{1}$$

or

$$PE_{\text{max}} = 1 - \phi_{\text{c}}\phi_{\text{f}} \tag{2}$$

where PE_c and PE_f are the packing efficiency of the coarse and fine particle fractions, respectively, and $\phi = 1 - PE$, is the interstitial pore fraction of

"To whom correspondence should be addressed at Thermo-Disc, 1981 Port City Boulevard, Muskegon, MI 49443, USA. *Maximum packing density is used in a macroscopic sense; particles are assumed to be randomly arranged. The term packing density and packing efficiency are used interchangeably. packed particles of a single size (fine or coarse particles only). Notice that the ratio of two terms of the right side of Eqn (1) is the volume ratio of the coarse to the fine particles to achieve the maximum packing efficiency.

The packing mechanism of the Furnas model is such that smaller particles are introduced and distributed to the interstices of larger packed particles so that the porosity is reduced. The calculation of the ideal packing density is based on the assumption that the size ratio of coarse particles to fine particles is infinitely large. It has been observed in practice by many researchers that when this particle size ratio decreases, the packing efficiency of mixed powders is lower than that predicted by the Furnas model (the Westman-Hugill diagram, Fig. 1). In order to understand the reason for those packing density deviations, Messing and Onoda 15,16 developed a packing model on the notion that the Furnas model may apply locally, but the different volume fractions of coarse and fine particles through the mixture lead to reductions in the overall packing density. Zok and Lange¹⁷ recently studied the effects of the surface of inclusions (the coarse particles) and the contacts of inclusions on the overall packing density. This theoretical analysis was on the scale of the dimension of the fine particles. However, the results from that study are not convenient for the calculation of packing density of a binary particle system with a variable volume fracion, particle size ratio, and different initial packing efficiencies.

Various empirical equations have been developed to predict the particle packing density of a binary system, ^{18–21} Some equations are either too complicated to apply, ¹⁸ or only consider the initial packing efficiency as a constant. ¹⁹ Most work in this research field has been reviewed recently. ²²

In this paper, an empirical equation is developed, which takes into account the particle size ratio, the volume fraction of powders, and their initial packing efficiencies. Constants in the proposed equation are adjusted to fit experimental data, which makes predicting the packing density of the mixture more accurate.

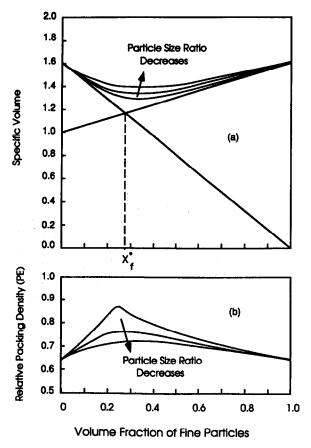


Fig. 1. The Furnas model. Theoretical variation of packing volume for the packing of smaller particles among coarser particles. (a) The Westman-Hugill plot of the Furnas model; straight lines correspond to ideal packing when the size ratio is infinite, and curved lines indicate typical behavior. (b) The Furnas model plotted as the packing density versus particle volume fraction. Figure 1(a) is preferred because the straight lines are easy to draw and their physical meaning is apparent. $X_{\rm f}^{\star}$ is the volume fraction of fine particles to achieve the maximum packing density.

Derivation of the Equation

For a coarse-fine binary particle size composite, the maximum packing density is predicted by the Furnas model (eqn (1)). The terms on the right side of eqn (1) are the volume fractions of coarse and fine particles, respectively. Starting with coarse powders and adding fine powders, the initial packing density is equal to the first term: the second term indicates the maximum possible density increase when the fine powder is added. For a real binary mixture, the maximum packing density increase should also be a function of the volume fraction and the size ratio of coarse to fine particles. Hence, we propose the following equation to calculate the packing density of a binary mixture (PE_{mix}) :

$$PE_{\text{mix}} = PE_c + (1 - PE_c)PE_f F_1(X_f) F_2(R)$$
 (3)

where F_1 is a function of the volume fraction of fine particles, X_6 ; F_2 is a function of the size ratio

Table 1. Volume fractions of coarse and fine particles calculated from the theoretical Furnas model

Packing efficiency $PE_{c} = PE_{f}$	Volume fraction	
	X _c	$X_{\mathrm{f}}^{\bigstar}$
0.6	0.714	0.286
0.5	0.667	0.333
0.4	0.625	0.375

of coarse particles to fine particles, R (average particle size ratio if the pure coarse or fine particles are not exactly monosize particles). It is seen, from Fig. 1, that F_1 must be zero when $X_f = 0$ or 1, and F_1 would reach its maximum when $X_f = X_f^*$ (X_f^* is the volume fraction of the fine particles at which maximum packing density is achieved). Figure 1 also indicates that F_2 must be 0 when the size ratio is 1, and F_2 is 1 when the size ratio is infinite.

For solutions to eqn 3 the functions F_1 and F_2 must be assumed or determined.

Selection of F_1

To select F_1 , a realistic X_f^* value must first be determined. Table 1 lists the theoretical value of X_f^* calculated from the Furnas model with the assumption that $PE_c = PE_f$. For spherical-like monosize particles, the packing efficiency for random packing is around 0.6; and hence, from Table 1, $X_f^* \approx 0.3$. Table 1 shows that X_f^* increases slightly with a decrease of the packing efficiency value for monosize particles. We have also observed that measured data from previous research^{1,2} show that as the particle size ratio decreases, the X_f^* value tends to increase slightly. In most cases, the X_f^* is between 0.3 to 0.4.

Bearing this value in mind, F_1 is arbitrarily chosen as:

$$F_1(X_f) = |e| X_f \ln X_f|^{C_1}$$
 (4)

where e is the base of a natural logarithm and C_1 is a constant. F_1 versus X_f with different C_1 values are plotted in Fig. 2. It is seen from Fig. 2, that the curve of this equation meets the required criteria, i.e. it is zero when X_f equals 0 or 1, and reaches the maximum when X_f is between 0.3 to 0.4.

It is also seen from Fig. 2, that the constant C_1 affects the shapes and the curvature of the curves. The procedure to determine C_1 is to assign different C_1 values into function F_1 ; then substitute F_1 and F_2 into eqn (3) and use the data from eqn (3) to plot the Westman-Hugill diagram. The C_1 giving the best match to the experimental data is then selected.

After comparing C_1 values for different binary systems, it was found that by taking C_1 as a function

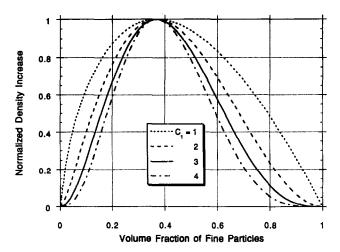


Fig. 2. Normalized packing density variation F_1 as a function of volume fraction of fine particles, X_f . F_1 is zero when X_f equals 0 or 1; F_1 is maximum when X_f is between 0.3 to 0.4. Value of the constant, C_1 , alters the shape of the curves.

of PE_c , instead of a constant, a better fit was obtained. The function is:

$$C_1 = \frac{5}{4PE_c} \tag{5}$$

The fact that C_1 is a function of PE_c is actually expected because the curves in the Westman-Hugill diagram are slightly different for binary systems with different PE_c and PE_f values. The Westman-Hugill data approaches the theoretical maximum packing density, the straight lines in Fig. 1(a), when the particle size ratio is infinitely large. When the particle size ratio is small, the data is continuous. In Fig. 2, smooth curves for F_1 show that the empirical equation derived in this paper is only suitable for systems with small particle size ratios. A particle size ratio of 10 or lower is considered small. If the particle size ratio is larger than 10, the packing density of the system is more closely predicted by the theoretical model.

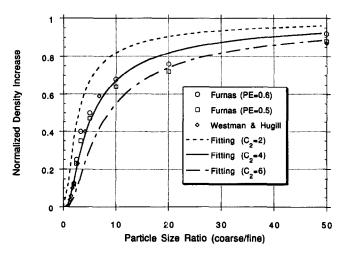


Fig. 3. Normalized packing density variation as a function of the articles size ratio for experimentally measured data and model. $C_2 = 4$ for best fit to measured data.

Selection of F_2

Function F_2 governs the increase of packing density as a function of the particle size ratio. It is seen that the maximum increase in packing density by adding fine particles to coarse, $PE_f(1 - PE_c)$, is a function of the initial packing efficiencies of coarse and fine particles. In order to compare the effects of particle size ratio on the increase in packing density, F_2 is normalized by $PE_f(1 - PE_c)$. Hence, F_2 is usually less than 1 for a real binary powder system. Figure 3 shows the normalized maximum packing density increase versus the particle size ratio of previous researchers. According to the distribution of these data, the fitting function F_2 is proposed as:

$$F_2 = \exp(\frac{-C_2}{R}) \tag{6}$$

where C_2 is a constant. It is seen, from Fig. 3, that a smaller C_2 leads to a more rapid increase of the F_2 value when the particle size ratio increases. It is also seen that when C_2 equals 4, the curve most closely fits the measured data.

After empirically obtaining the functions of F_1 and F_2 , eqn (3) is combined with eqns (4), (5) and (6), to get the full descriptive equation for calculating the packing density of a binary mixture:

$$PE_{\text{mix}} = PE_{\text{c}} +$$

$$(1 - PE_{\text{c}})PE_{\text{f}}[e \cdot X_{\text{f}} \cdot \ln X_{\text{f}}]^{\frac{5}{4PE_{\text{c}}}} \exp\left(-\frac{4}{R}\right)$$
(7)

The specific volume of binary mixtures calculated from eqn (7) is plotted in Figs 4 and 5. The calculations are based on the assumption that in the binary mixtures $PE_c = PE_f$, and PE_f equals 0.6 and 0.5, respectively for Figs 4 and 5. It is observed from Figs 4 and 5, that the curves

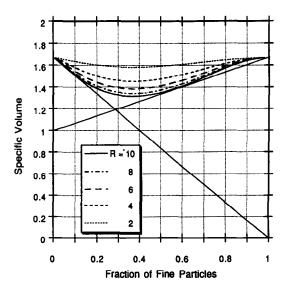


Fig. 4. Calculated specific volume versus particle size ratio using eqn (7). Initial packing efficiency $PE_c = PE_f = 0.6$.

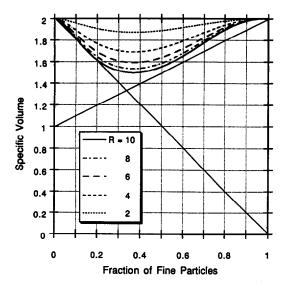


Fig. 5. Calculated specific volume versus particle size ratio using eqn (7). Initial packing efficiency $PE_c = PE_f = 0.5$.

calculated by this empirical equation are generally similar to what is observed in practice, except that at a high volume fraction of fine particles, the calculated specific volume is slightly higher than that seen in practice. For example, if the volume fraction of fine particles is 0.8–0.9, in Fig. 5, the calculated packing density by eqn (7) could be about 2% lower than the measured one.

For a binary powder mixture $PE_c \neq PE_f$

For a binary powder system having $PE_c \neq PE_f$, eqn (7) can still be used when the volume of the coarse powders is dominant in the mixture. For a system in which the volume of fine particles is dominant, PE_f may be used as the first term in eqn (7), instead of PE_c . Figure 6 shows the experimentally measured specific volume, as well as the

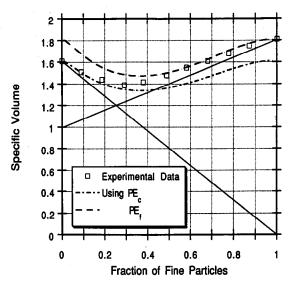


Fig. 6. Calculated specific volume versus particle size ratio using both PE_c or PE_f in the first term of eqn (7) when PE_c is not equal to PE_f .

values calculated by eqn (7), using both PE_c or PE_f as the first term. This shows that when the volume fraction of fine particles is less than 0.3, the calculated data using PE_c fits the experimental data. When the volume fraction of fine particles is larger than 0.5, the calculated data using PE_f fits the experimental data. If the volume of fine particles is between 0.3 to 0.5, the experimental data is between the two calculated values.

Summary and Conclusion

The Furnas model describes the ideal particle packing behavior of a binary powder system. The packing density, however, is lower when the size ratio of coarse particles to fine particles decreases. Packing density also depends on the volume fraction of coarse or fine particles. An empirical equation (eqn (7)) was derived to estimate the packing density of a real binary system. This equation, which takes into account the particle size ratio, the compositional variations of the system, and the initial packing efficiencies of particles as variables, enables one to easily predict the packing density of a binary particle system from its particle size ratio and volume fractions of coarse and fine particles.

References

- Furnas, C. C., The relations between specific volume voids, and size composition in system of broken solids of mixed size. In Report of Investigations, Department of Commerce-Bureau of Mines, No. 2894, Oct., 1928.
- Westman, A. E. R. & Hugill, H. R., The packing of particles, J. Am. Ceram. Soc., 13[10] (1930) 767-9.
- 3. Furnas, C. C., Grading aggregates, I—mathematical relations for beds of broken solids of maximum density, *Industrial and Engr. Chem.*, 23[9] (1931) 1052-8.
- Anderegg, F. O. Grading Aggregates, II—The application of mathematical formulas to mortars, *Industrial and Engr. Chem.*, 23[9] (1931) 1058-64.
- Andreasen, A. H. M. & Andersen, J., Relation between grain size and interstitial space in products of unconsolidated granules, Kolloid-Z., 50 (1930) 271.
- Schuhmann Jr., R., Principles of comminution: I, size distribution and surface calculation, *Mining Tech.*, 4[4] (1940) 1-11.
- 7. McGeary, R. K., Mechanical Packing of Spherical Particles, J. Am. Ceram. Soc., 44[10] (1961) 513-22.
- Sohn, H. Y. & Moreland, C., The effect of particle size distribution on packing density, Can. J. Chem. Eng., 44, (1968) 162-67.
- Bierwagen, G. P. & Saunders, T. E., Studies of the effects of particle size distribution of the packing efficiency of particles, *Powder Tech.*, 10 (1974) 111-19.
- Fedors, R. F. & Landel, R. F., An empirical method of estimating the void fraction in mixtures of uniform particles of different size, *Powder Technology*, 23 (1979) 225-31.
- 11. Patankar, A. N. & Mandel, G., The packing of solid particles, *Trans. Indian Ceram. Soc.*, 39[4] (1980) 109-117.

- 12. Lee, D. I., Packing of spheres and its effect on the viscosity of suspensions, J. Paint Tech., 42[11] (1970) 579-87.
- 13. Funk, J. E., Dinger, D. R. & Funk, Sr., J. E., Control parameters for a 75 wt% coal-water slurry: Co-Al. In Proceedings of the 4th International Symposium on Coal-Slurry Combustion, Pittsburgh Energy Technology Center/DOE, Pittsburgh, PA, 1982.
- Zheng, J., Johnson, P. F. & Reed, J. S., Improved equation of the continuous particle size distribution for dense packing, J. Am. Ceram. Soc., 73[5] (1990) 1392–8.
- Messing, G. L. & Onoda, Jr, G. Y., Inhomogeneity-packing density relations in binary powders, J. Am. Ceram. Soc., 61[1-2] (1978) 1-5.
- Messing, G. L. & Onoda, Jr, G. Y., Inhomogeneity-packing density relations in binary powders Experimental Studies, J. Am. Ceram. Soc., 61[7-8] (1978). 363-6.

- Zok, F., Lange, F. F. & Porter, J. R., Packing density of composite powder mixtures, J. Am. Ceram. Soc., 74[8] (1991) 1880-5.
- 18. Westman, A. E. R., The packing of particles: empirical equations for intermediate diameter ratios, *J. Am. Ceram. Soc.*, 19 (1936) 127-9.
- 19. Yerazunis, S., Cornell, S. W. & Winter, B., Dense random packing of binary mixtures of spheres, *Nature*, **207**[8] (1965) 835–7.
- Fedors, R. F. & Landel, R. F., An empirical method of estimating the void fraction in mixtures of uniform particles of different size, *Powder Tech.*, 23 (1979) 225-31.
- Yu, A. B. & Standish, N., Porosity calculation of multicomponent mixtures of spherical particles, *Powder Tech.*, 52 (1987) 233-41.
- German, R. M., Particle Packing Characteristics, Metal Powder Industries Federation, Princeton, New Jersey, 1989.