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VOLUME CONTRACTION IN LIQUID FLUIDIZATION OF BINARY SOLIDS MIXTURES

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ABSTRACT

When liquid-fluidized particles of radically different sizes and densities mix, the serial (additive volume) model fails to predict the fluidized bed voidage due to contraction of the mixed bed relative to the volumetric sum of the corresponding monocomponent beds. Models are proposed to predict contraction reported in the literature for both upflow and downflow fluidization.

INTRODUCTION

This study concerns liquid fluidization of binary solids, i.e. mixtures containing two particle species, where a species is defined as a collection of solid particles of uniform size, shape and density. When the two species differ only in size, or only in shape (Escudié et al., 1) or even moderately both in size and density, the fluidized mixture at any given liquid velocity usually follows what has become known as the serial model (Epstein et al., 2). According to this model, the volume occupied by the fluidized bed binary is the sum of the volumes occupied by monocomponent beds of the two constituent particle species, each fluidized at the same superficial liquid velocity as for the binary. The model applies irrespective of particle mixing caused by flow instabilities that over-ride the bulk density differences which would otherwise cause the two particle species to segregate. If, however, a binary consists, say, of fixed shape particles, e.g. spheres, having a relatively large diameter ratio in excess of unity, but a corresponding buoyancy-modified density ratio far below unity, then the possibility arises that the two species may form a bulk-density-balanced equilibrium mixture which is independent of any flow instabilities (Escudié et al., 3). The bottom mixed layer that occurs in the progression of the well-studied layer inversion phenomenon for conventional upflow liquid fluidization (Escudié et al., 4) is an example of such an equilibrium mixture, which encompasses the entire bed at the layer inversion point. In the case of downflow (inverse) liquid fluidization, the mixed layer is located at the top of the column, close to the distributor. For these mixed layers, and especially at or near the inversion point, Chiba (5) and Asif (6, 7) for upflow fluidization and Escudié et al. (8) for downflow, have shown that the serial model fails to predict the layer (or bed) voidage, due to a significant contraction relative to the corresponding monolayers in series. This contraction is the subject of the present investigation.

EXPERIMENTAL STUDIES

Table 1 summarizes the data selected from the literature. Species *1* refers to the larger particles and species *2* to the smaller particles. In conventional upflow fluidization, species *1* also denotes the lower-density particles and species *2* the higher-density particles, whereas it is the reverse for downflow fluidization.

Six studies covering eleven binary combinations have been selected for upflow fluidization. To investigate the bed contraction of the mixed layer generated during the inversion progression (with or without a pure monocomponent layer), it is necessary to obtain information that was not always reported: (i) Richardson and Zaki (9) expansion index, n_i, and extrapolated intercept, U_{ti}, for the monocomponent beds of each particle species; (ii) both the voidage and the solids composition of the mixed layer for a given superficial liquid velocity, U. More papers have been published on inversion in conventional fluidization than the six selected here (Escudié et al., 4), but these could not be used because one or more of the above was not reported. As the serial model is very sensitive to n_i and U_{ti}, only directly measured values were used. For the binaries from the literature, the size ratio, d_1 / d_2, ranged from 2.19 to 10.0. Most particles were spherical, the only exception being species *1* of Asif (7), which had a sphericity of 0.85.

In the case of downflow fluidization, the regime designated as incomplete segregation by Escudié et al. (1) was observed for high liquid velocities, a monocomponent layer of smaller particles being located at the bottom of the column (far below the distributor), whereas a mixed layer of species appeared at the top, closer to the distributor. The voidage and solids composition of the mixed layer were estimated for two binary combinations. In addition, a "heterogeneous mixing" pattern (Escudié et al., 1) was also observed for the lower superficial liquid velocities. In this regime, the fluidized bed consists of only one mixed layer, its overall liquid-free composition being equal to the liquid-free volume fraction of both particle species in the bed. Two sets of data, based on the mixed layer in the incomplete segregation regime and in the heterogeneous mixing regime, were thus included in the present study.

Table 1. Characteristics of the binaries included in this work, and number of available experimental data points

		Particle properties				Experimental Richardson-Zaki parameters				No. of overall solids compositions	No. of exp. voidages
		d_1 mm	ρ_1 kg/m^3	d_2 mm	ρ_2 kg/m^3	U_{t1} m/s	U_{t2} m/s	n_1	n_2		
Escudié et al. (9)	Bin. I	15.0	895.9	6.35	836.5	0.167	0.131	3.08	2.82	4	39 + 27*
	Bin. II	19.1	903.0	6.35	836.5	0.171	0.131	2.43	2.82	3	19 + 17*
Moritomi et al. (10) Chiba (5)	Bin. I	0.775	1380	0.163	2450	0.0465	0.0143	3.00	4.01	5	28
	Bin. III	0.385	1380	0.163	2450	0.0148	0.0143	2.56	4.01	1	1
Jean and Fan (11)		0.778	1509	0.193	2510	0.0370	0.0175	3.85	4.25	3	3
Matsuura and Akehata (12)		2.01	1170	0.807	1395	0.0592	0.0462	3.28	3.51	5	41
Funamizu and Takakuwa (13)	Bin. I	0.302	1418	0.091	2473	0.0151	0.0059	3.83	4.35	1	12
	Bin. II	0.486	1386	0.134	2476	0.0244	0.0130	3.65	3.87	5	17
	Bin. III	0.677	1398	0.134	2476	0.0380	0.0130	3.31	3.87	1	19
	Bin. IV	0.677	1398	0.164	2480	0.0380	0.0177	3.31	3.77	1	23
Asif (7)	Bin. I	2.76	1396	0.275	2664	0.0937	0.0346	2.61	3.79	6	6
Rasul (14)	Bin. I	0.855	1360	0.138	2920	0.0461	0.0154	3.65	4.16	1	44
	Bin. II	0.181	2450	0.0825	4200	0.0191	0.0123	4.17	4.69	1	8

* data corresponding to "heterogeneous mixing" pattern.

Table 1 summarizes the data for each binary. The total number of data points is 304, of which 202 and 102 correspond to upflow and downflow fluidization, respectively. The liquid was water at room temperature and pressure in all cases.

RESULTS

Volume contraction

The percentage volume change (which usually turns out to be negative, denoting contraction) from the serial model can be calculated by comparing the experimental voidage ε_{exp} to the value, ε_{serial}, estimated from the serial model, as follows:

$$\text{Volume change (\%)} = \left(\frac{1/(1-\varepsilon_{exp}) - 1/(1-\varepsilon_{serial})}{1/(1-\varepsilon_{serial})} \right) \times 100\ \% \qquad (1)$$

where ε_{serial} is the voidage predicted by the serial model (Epstein et al., 2), i.e. by

$$\frac{1}{1-\varepsilon_{serial}} = \sum_{i=1}^{2} \frac{x_i}{1-\varepsilon_i} \qquad (2)$$

ε_i being the monocomponent voidage at the given U for the ith particle species and x_i the fluid-free volume fraction of particle species i in the mixed layer.

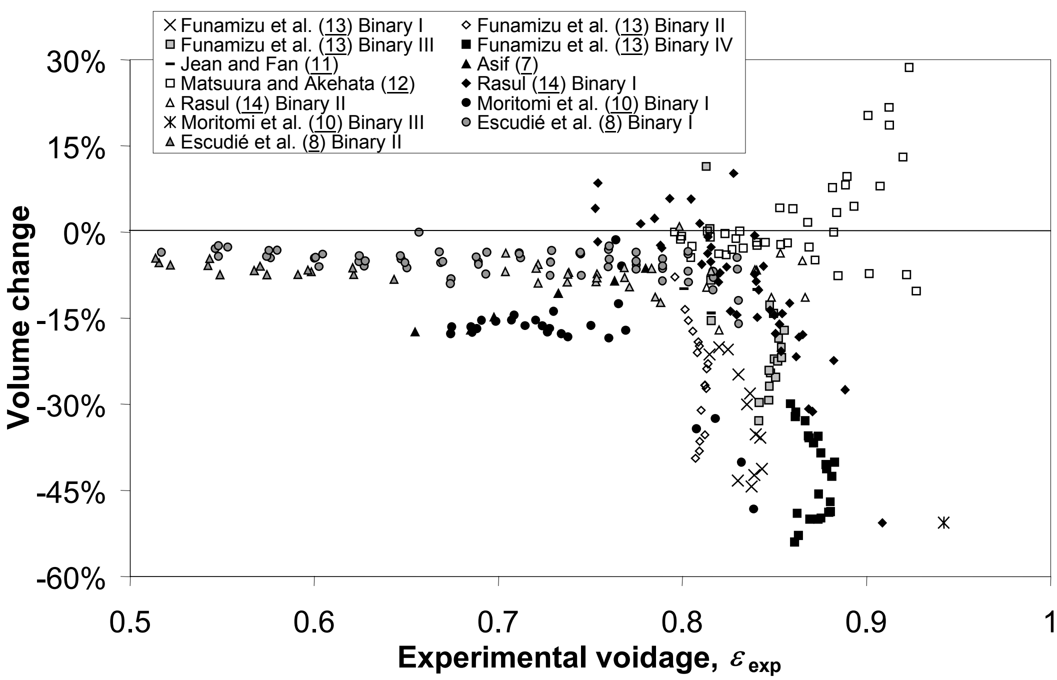

Figure 1. Volume change, as defined by Eq. (1), of the binary mixtures selected for this study as a function of the experimental voidage.

Fig. 1 plots the volume change of the data against the experimental voidage. Most points range between 0 and –20%. However, the maximum magnitude reaches about –50% for five binaries: from binaries I and III of Moritomi et al. (10), binaries I and IV of Funamizu and Takakuwa (13), and binary I of Rasul (14). Some points also show “volume expansion” compared to the serial model, particularly the data of Matsuura and Akehata (12). In that study, the solids hold-up of each species was deduced from visual measurements of the total height of the fluidized bed with binary particles and from the visual height of the mixed layer. Even if each species had a narrow particle size distribution, the boundary between the upper and lower layers was less distinct than the top surface of the fluidized bed. The boundary was taken at the average mid-point of the maximum and minimum heights of the transition (fuzzy interface) region, which is usually thin. The uncertainty of the boundary can affect the quality of the voidage estimation, and it is therefore not surprising that the "bed expansion" corresponds to the highest superficial liquid velocities where the boundary was especially indistinct.

The influence of the voidage (and the related superficial liquid velocity) on the contraction of each binary system is not clear. Even if contraction intensifies with increasing voidage for the four binaries of Funamizu and Takakuwa (13) and binary I of Rasul (14), this effect is not observed for binary II of Rasul (14), nor for those of Jean and Fan (11) and the two of Escudié et al. (8). For the mixture of Asif (7), an increased voidage even had a negative effect on the contraction.

Voidage prediction

Several schemes for predicting the contraction effect have been proposed. The simplest, that of Gibilaro et al. (15), treats the expansion of a binary mixture as if it were a monocomponent bed with a Sauter mean diameter and a volumetric mean density of the two particle species. Developed to estimate the composition of the mixed layer, this method of implicitly incorporating a contraction effect was used in their "complete segregation model" of the layer inversion phenomenon in conventional fluidized beds. The same method, but with a different bed expansion equation, that of Di Felice (16), was explored by Epstein (17). Other models based on a similar approach were developed subsequently, e.g. combining the Richardson and Zaki (9) equation with a property-averaged terminal velocity $\overline{U_t}$ and index $\overline{n}$ (Asif, 18; Escudié et al., 4). However, these models are handicapped by having to make their predictions from calculated rather than from measured values of the monocomponent voidages.

An entirely different method, initiated by Asif (6), is based on the packing of binary mixtures of spheres. These models utilize the equation of Westman (19),

$$\left(\frac{V - V_2 x_2}{V_1}\right)^2 + 2G\left(\frac{V - V_2 x_2}{V_1}\right)\left(\frac{V - x_2 - V_1(1-x_2)}{V_2 - 1}\right) + \left(\frac{V - x_2 - V_1(1-x_2)}{V_2 - 1}\right)^2 = 1 \qquad (3)$$

where $V = (1 - \varepsilon)^{-1}$, $V_i = (1 - \varepsilon_i)^{-1}$ and the dimensionless parameter G incorporates the contraction effect, being unity for the serial model (no contraction).

The principal challenge of this procedure is the estimation of G. Empirical equations have been proposed in the packed bed literature. According to Yu et al. (20), G is only a function of the diameter ratio of the binary spheres:

$$G = \left(a r^b\right)^{-1} \text{ for } r \le 0.824\text{ ; } G = 1 \text{ for } r \ge 0.824 \qquad (4)$$

where $r = d_2 / d_1$ is the size ratio, a = 1.355 and b = 1.566. Finkers and Hoffman (21) included $(1 - \varepsilon_1)^-$ and $(1 - \varepsilon_2)$ as additional variables in their more complicated empirical equation for G applied to non-spherical particles:

$$G = r_{str}^k + \left(1 - \varepsilon_1^{-k}\right) \qquad (5)$$

where

$$r_{str} = \left(\varepsilon_1^{-1} - 1\right) r^3 \big/ \left(1 - \varepsilon_2\right) \qquad (6)$$

and $k = -0.63$.

Table 2. Accuracy of the models predicting the parameter *G* in Eq. (3)

Model	Equation number	No. of fitted parameters	Parameter values	*AAD* %
1. G = constant		1	$G = 2.17$	2.48
2. $G = \text{fn}(k)$	5 and 6	1	$k = -0.340$	2.31
3. $G = \text{fn}(r)$	4	2	$a = 5.31$ $b = 2.48$	1.92
4. $G = \text{fn}(r, Ar_1/Ar_2)$	9	3	$c = 2.65$; $d = 0.995$ $e = -0.766$	1.83
5. $G = \text{fn}(r, Ar_1, Ar_2)$	10	4	$f = 7.69$; $g = 12.2$ $h = -0.239$; $i = 0.246$	1.67

Increasing the complexity of the correlation to estimate G or the number of fitted parameters may improve the volume contraction prediction. Here we test:

1. Single value of G for all binary systems (one-parameter model);
2. Equations (5) and (6), but with k as a fitting parameter;
3. Equation (4), but with both a and b fitted;
4. A correlation that accounts for the particle density ratio in addition to the diameter ratio,

$$G = \left(c\, r^{d} \rho^{e}\right)^{-1} \tag{7}$$

The density ratio, $\rho = (\rho_2 - \rho_f)/(\rho_1 - \rho_f)$, which is positive and greater than unity for both upflow and downflow fluidized beds, can be rewritten using the Archimedes number of each particle species,

$$Ar_i = d_i^3 \rho_f \left(\rho_i - \rho_f\right) g / \mu^2 \tag{8}$$

Eq. (7) can thus be expressed as

$$G = \left(a\, r^{d-3e} \left(Ar_2/Ar_1\right)^{e}\right)^{-1} \tag{9}$$

5. A four-parameter model taking into account the size ratio and Archimedes number of each particle species,

$$G = \left(f\, r^{g} \left|Ar_1\right|^{h} \left|Ar_2\right|^{i}\right)^{-1} \tag{10}$$

All five models for predicting the parameter G were tested. The number of fitted parameters ranges from one to four. To determine quantitatively the accuracy of the predictions, we have calculated an average absolute % deviation, AAD:

$$\text{AAD} = \sum_{i=1}^{N} \left| \frac{\varepsilon_{\text{predicted}} - \varepsilon_{\text{exp}}}{\varepsilon_{\text{exp}}} \right|_i \cdot \frac{100}{N} \tag{11}$$

where $N = 304$ is the number of data points. Table 2 presents, for these models, the best-fitting values of the parameters and their respective AAD values. By way of comparison, AAD calculated for the serial model is 3.43%. It is not surprising to observe that the agreement between the predicted and experimental voidages improves with an increase in the number of fitted parameters: AAD is about 2.3-2.5% for the one-parameter models, 1.92% for the two-parameter model and 1.67% for the four-parameter model. Figure 2a plots the experimental voidage against the predicted voidage from the four-parameter model. The gains in voidage estimation relative to the serial model are particularly significant for the binary of Asif (7), binary II of Funamizu and Takakuwa (13), and most data for binary I of Moritomi et al. (10).

The relevance of the Westman (19) equation for predicting the volume change is now investigated. Four simplified models were tested to estimate the volume change directly.

a) Constant extent of volume contraction.

b) Since the particle diameter ratio influences the degree of contraction,

$$\text{Fractional volume change} = a\left(1 - r^{b}\right) \quad (12)$$

c) Since the prediction of G was improved by accounting for the Archimedes number ratio of the particle species,

$$\text{Fractional volume change} = c\left(1 - r^{d}\left(Ar_2/Ar_1\right)^{e}\right) \quad (13)$$

d) A fourth model similar to Eq. (10) was also defined with

$$\text{Fractional volume change} = f\left(1 - r^{g}\left|Ar_1\right|^{h}\left|Ar_2\right|^{i}\right) \quad (14)$$

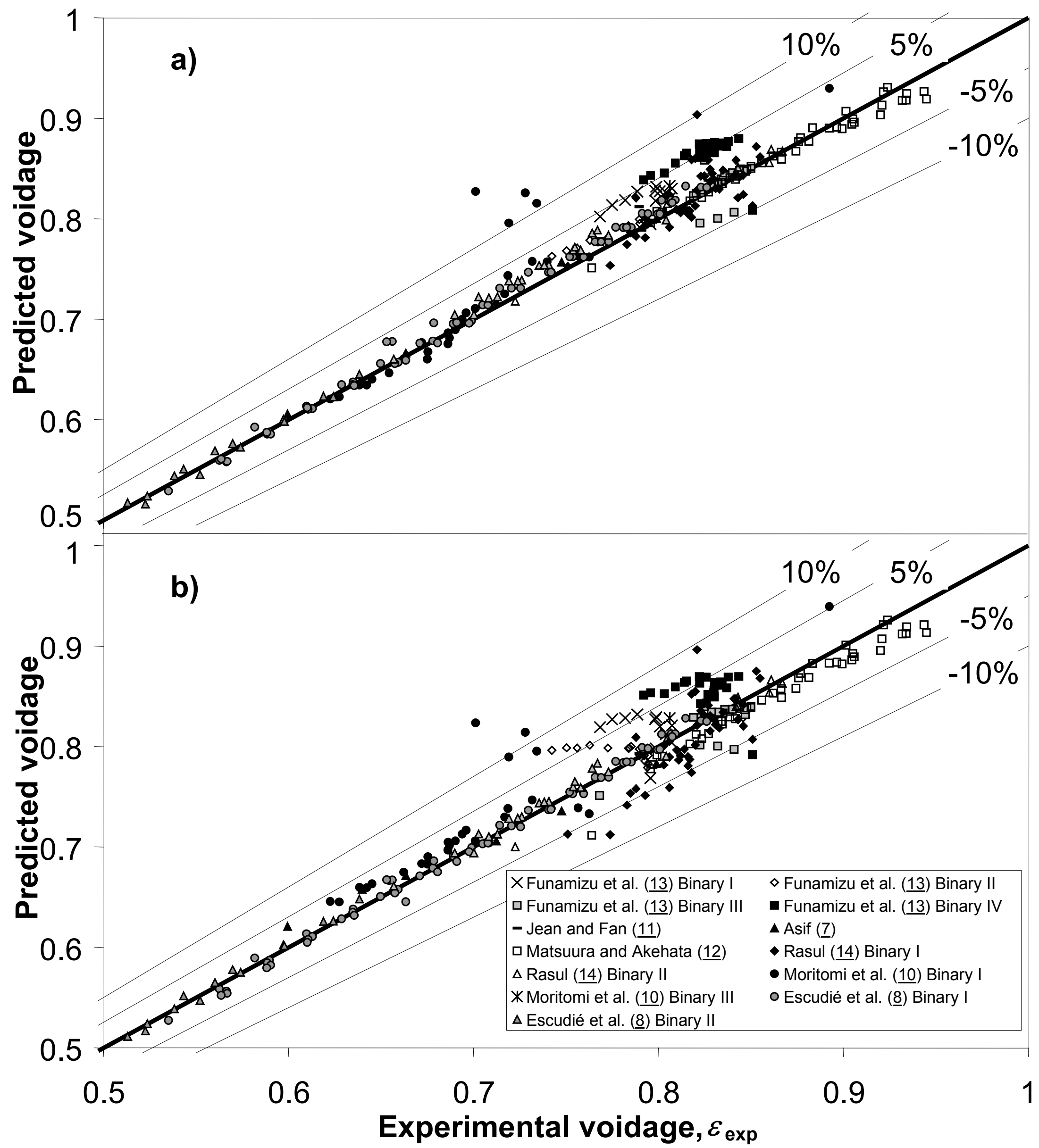

Figure 2. Comparison of experimental voidage with predictions: a) Four-parameter model using Eq. (10) for estimation of the parameter G in Eq. (3); b) Four-parameter model using Eq. (14) based on direct estimation of the volume contraction.

Table 3. Accuracy of models predicting directly the volume change

Model of volume change	Equation number	No of fitted parameters	Parameter values	*AAD* %
a) constant		1	-0.0601	2.23
b) fn(r)	12	2	a = -1.86; b = 0.0338	2.03
c) fn (r,Ar_1/Ar_2)	13	3	c = -0.279; d = 1.29 e = -0.427	1.94
d) fn (r,Ar_1,Ar_2)	14	4	f = -0.373; g = 0.9604 h = 0.3254; i = -0.3234	1.94

Table 3 summarizes the accuracy of these four models. Figure 2b presents the voidage calculated from the four-parameter model (Eq. 14). Although the accuracy of the estimations is better for a one-parameter model when the volume contraction is calculated directly rather than via G, the models based on the Westman equation do better as the number of parameters increases. As shown in Fig. 2b, the four-parameter model overestimates by more than 5% some data (binaries I, II and IV of Funamizu and Takakuwa, 13; six data for binary I of Rasul, 14), and also underestimates by less than -5% seven data for binary I of Rasul (14), and one each for binaries III and IV of Funamizu and Takakuwa (13) and that of Matsuura and Akehata (12). Comparison of Figs. 2a and 2b shows the advantage of basing a multi-parameter model on the Westman equation.

CONCLUSIONS

When particles of radically different sizes mix homogeneously or even heterogeneously, which occurs most strikingly under some conditions where the size ratio and the ($\rho_i - \rho_f$) ratio of the two particle species are on opposite sides of unity, the serial model fails due to contraction of the mixed bed relative to the volumetric sum of the corresponding monocomponent beds. Such contractions have been reported in conventional upflow liquid fluidization for six studies covering eleven binary combinations. These results are supplemented here by our own experimental data on downflow fluidization of two binary combinations.

Two approaches were developed to predict the voidage: correlations based on direct estimation of the volume contraction, and correlations to estimate the parameter G in the Westman (19) equation. In both cases, the correlations contained anywhere from one to four adjustable parameters, accounting variously for the particle diameter ratio and the Archimedes numbers of each particle species. The accuracy of the estimations is better for the models based on the Westman equation, by which the voidage could be predicted with good accuracy (±1.67%) using four fitted parameters.

NOTATION (for terms not explicitly defined in the text)

d diameter of solid spheres, mm or m
g gravitational acceleration, m.s^{-2}
U_t value of U when a linear plot of log U vs. log ε is extrapolated to ε = 1, m.s^{-1}
ε overall voidage, dimensionless
μ liquid viscosity, kg.m^{-1}.s^{-1}
ρ_f liquid density, kg.m^{-3}
ρ_i density of solid particle species i, kg.m^{-3}

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