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1 **Comparison of the properties of segregated layers in a bidispersed**  
2 **fluidized bed to those of a monodispersed fluidized bed**

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## Abstract

17

18 Since industrial fluidized-bed reactors typically operate with polydispersed particles, the abil-  
19 ity to approximate such reactors as the superposition of corresponding monodispersed fluidized  
20 beds would greatly simplify their design and operation. To evaluate the validity of superposition  
21 of monodispersed reactor behavior, we evaluate the effects of bidispersity by comparing three-  
22 dimensional liquid-solid monodispersed and segregated bidispersed fluidized beds. Simulations  
23 were conducted using the Immersed Boundary Method (IBM) with direct forcing in a periodic  
24 domain and with particle Reynolds numbers of 20-70 based on the largest particle diameter. We  
25 show that the volume fraction, kinematic wave speed, particle velocity fluctuations, and collisional  
26 and hydrodynamic stresses in the segregated layers of a bidispersed fluidized bed can be well ap-  
27 proximated by the corresponding properties of a monodispersed fluidized bed. In the transition  
28 region between the layers, only the volume fraction and collision stresses monotonically decrease  
29 with height. At low Reynolds numbers, particle velocity fluctuations in the upper layers are the  
30 largest. As the particle Reynolds number increases, particle velocity fluctuations in the transition  
31 and lower layers become the largest sequentially. At intermediate particle Reynolds numbers, the  
32 hydrodynamic stresses in the transition region are greater than those in the upper and lower layers.  
33 As the particle Reynolds number increases, the difference between the hydrodynamic stresses in  
34 the transition layer and the two layers becomes more significant. This work demonstrates that,  
35 despite the clear segregation into layers that behave like monodispersed beds, the transition region  
36 is governed by complex bidispersed mechanisms that cannot be explained in terms of the parti-  
37 cle behavior in the segregated layers. Overall, particle dynamics of the segregated layers in the  
38 bidispersed fluidized bed can be approximated with the corresponding monodispersed layers. The  
39 result implies that industrial applications such as wastewater treatment performance in bidispersed  
40 or polydispersed fluidized beds can be predicted with results from past numerical or experimental  
41 studies of monodispersed fluidized beds.

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## 42 I. INTRODUCTION

43 Liquid-solid fluidization is found in many industrial systems such as wastewater treatment  
44 and chemical processes. In wastewater treatment, fluidized-bed reactors are widely used in  
45 treating both industrial and domestic wastewater [1–3]. Traditional biological domestic  
46 wastewater treatment is energy intensive [4], leading to the development of the Staged  
47 Anaerobic Fluidized-bed Membrane Bioreactor (SAF-MBR) that reduces energy demand in  
48 wastewater treatment by recovering energy in the form of methane [2, 5, 6]. In the SAF-  
49 MBR, wastewater is injected at the reactor bottom to fluidize granular activated carbon  
50 (GAC) which is used to support the growth of microorganisms. Like many other fluidized  
51 beds, the non-uniformity of the GAC particles introduces more complexity in the fluidized-  
52 bed hydrodynamics. The ability to predict the hydrodynamics of fluidized beds used in  
53 wastewater treatment improves existing biological models for predicting and optimizing the  
54 treatment performance.

55 Since the monodispersed spherical fluidized bed is the most simplified and idealized  
56 fluidized-bed reactor, it has been studied extensively in the past [7–10]. Root-mean-square  
57 (rms) particle velocity fluctuations have been found to vary from 10% to 170% of the upflow  
58 velocity [8, 10–12]. A series of papers focus on establishing a relationship between the upflow  
59 velocity and volume fraction. The most widely adopted relationship is based on the power  
60 law model which can be applied to both fluidization and sedimentation, and is given by

$$u^* = \frac{u_0}{w_{ref}} = k(1 - \phi)^n, \quad (1)$$

61 where  $u_0$  is the superficial or upflow velocity of the fluidized bed,  $w_{ref}$  is the settling velocity  
62 of a single particle in the domain of interest,  $k$  is a constant to correct for high volume  
63 fractions [8, 13–15],  $\phi$  is volume fraction and  $n$  is the expansion or power-law exponent.  
64 Peak rms fluctuations have been shown to occur at a volume fraction of 30% [10]. At this  
65 volume fraction, the force on the particles transitions from collision to flow dominant [16].

66 Since monodispersed fluidized beds are not found in industrial applications, understand-  
67 ing the effects of polydispersity is critical. The ability to approximate polydispersed fluidized  
68 beds as the superposition of corresponding monodispersed fluidized beds would greatly  
69 simplify their design and operation by enabling application of existing understanding of  
70 monodispersed fluidized beds, for which there is extensive literature (i.e., equation 1). A  
71 bidispersed liquid-solid fluidized-bed represents a level of complexity that is sufficient to

72 understand the basic effects of multiple particle sizes yet is not as computationally costly as  
73 a fully polydispersed system because particles with the smallest diameter dictate the grid  
74 resolution. Nevertheless, the number of parameters increases substantially in the study of  
75 a bidispersed bed. Because two interacting particles can have both different diameters and  
76 densities, the following three different cases are possible: (1) same diameter but different  
77 density, (2) same density but different diameter and (3) different diameter and density. For  
78 cases (1) and (2), particle segregation generally occurs, whereby the large (dense) particles  
79 sink to the bottom layer while the small (light) particles rise to the top layer. In a transition  
80 region between the two layers, both particles coexist but tend to move in opposite directions  
81 and the thickness of the transition region decreases with increasing particle diameter ratio or  
82 particle density ratio. Complete segregation is assumed to occur when the particle diameter  
83 ratio is greater than two [13]. The height of bidispersed fluidized bed is typically assumed to  
84 be the sum of the heights of the two monodispersed fluidized-bed layers, and this has been  
85 shown to be quite accurate in sufficiently large systems [13, 17]. Nevertheless, a large body  
86 of work has been devoted to developing models to predict the thickness of the transition  
87 region by solving the steady-state advection-diffusion equation in the axial  $z$  direction of the  
88 bed for particle  $i = 1$  of 2,

$$-D_i \frac{\partial \phi_i}{\partial z} = \phi_i u_{seg,i}, \quad (2)$$

89 where  $D_i$  is the dispersion coefficient,  $\phi_i$  is the volume fraction and  $u_{seg,i}$  is the segregation  
90 velocity [18–20]. These papers demonstrate that the volume fraction of the layers in a segre-  
91 gated bidispersed fluidized bed can be approximated as a superposition of two monodispersed  
92 fluidized-bed layers. However, the dispersion coefficient and segregation velocity needed to  
93 predict the volume fraction are largely based on fitting and heuristics without quantification  
94 of the underlying particle microstructure physics. In this paper, we study these physics to  
95 obtain a more quantitative understanding of the processes affecting the transition region in  
96 a bidispersed fluidized bed.

97 To study the detailed hydrodynamics of a fluidized bed, there are both experimental  
98 and simulation approaches. In addition to the obvious advantage of studying real reactors  
99 without the need for models or simulations, the advantage of experiments is the ability to  
100 test a large number of different particle parameters in a relatively short period of time. This  
101 approach has been widely adopted to study macroscopic properties such as modeling  $Re_t$

102 and  $n$  [7, 21] and understanding the effect of particle properties on the collision pressure [22].  
 103 However, quantification of local hydrodynamics and microstructure in a fluidized bed with  
 104 experiments is difficult [8], most notably because direct imaging of particles is only possible  
 105 for dilute suspension [23, 24]. In recent years, high-fidelity particle-resolved simulations  
 106 (PRS) have gained popularity as a reliable technique to accurately resolve fluid-particle  
 107 interactions. The approach is based on first principles and approximations are only needed  
 108 to model particle collisions. With PRS, because individual particle information can be  
 109 tracked over time, a more detailed examination of microstructure and local hydrodynamics  
 110 in a fluidized bed is possible. Recently, the PRS approach has been widely adopted for a  
 111 number of different problems, including extracting drag laws from arrays of particles [25–28]  
 112 and understanding the detailed physics of flow-particle interactions in fluidized beds and  
 113 particle suspensions [8–11, 29–31]

114 In this paper, we present PRS results of liquid-solid monodispersed and segregated bidis-  
 115 persed fluidized-bed reactors to gain a detailed understanding of the effects of bidispersivity  
 116 on the particle dynamics. A series of cases with different particle Reynolds numbers is  
 117 studied and the simulation results are used to validate the assumption of approximating  
 118 segregated fluidized beds as the superposition of two monodispersed fluidized beds.

## 119 II. NUMERICAL METHODOLOGY AND SIMULATION SETUP

### 120 A. Equations and discretizations

121 The governing Navier-Stokes equations are solved in a three-dimensional rectangular  
 122 domain containing an array of spherical particles. A source term,  $\mathbf{f}_{\text{IBM}}$ , based on the direct-  
 123 forcing IBM method is added to the incompressible Navier-Stokes equation to enforce no-slip  
 124 boundary conditions on the particle surfaces as

$$\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} = -\nabla p + \nu_f \nabla^2 \mathbf{u} + \mathbf{f}_{\text{IBM}}, \quad (3)$$

125 subject to continuity,  $\nabla \cdot \mathbf{u} = 0$ , where  $\mathbf{u}$  is the velocity vector and  $p$  is the pressure normal-  
 126 ized by the fluid density,  $\rho_f$ . Equation 3 is solved on a uniform collocated Cartesian grid.  
 127 Coupling between the momentum and pressure equations is achieved using the fractional-  
 128 step method proposed by Ref [32]. The advection term is discretized with the explicit,  
 129 three-step Runge-Kutta scheme described in Ref [33]. The viscous term is discretized with

130 the implicit Crank-Nicolson scheme to eliminate the associated stability constraint. The  
 131 Hypre library is used to solve the linear systems arising from the implicit discretization  
 132 of the viscous terms and the pressure-Poisson equation [34, 35]. To solve the interactions  
 133 between the fluid and the particles, the direct forcing approach first proposed by Ref [36]  
 134 and improved by Ref [37] is adopted. Collision models [38, 39] are used to simulate particle-  
 135 particle interactions when the separation distance between the particle surfaces is less than  
 136 two grid cells. A detailed description and validation of the method can be found in Ref [40].

## 137 B. Simulation setup

138 Three-dimensional simulations are conducted in the rectangular, doubly-periodic (x-  
 139 and y-directions) domain shown in figure 1. The particles have a constant density  $\rho_p =$   
 140  $1300 \text{ kg m}^{-3}$ . Three different particle diameters  $d_{p,1}$ ,  $d_{p,2}$  and  $d_{p,3}$  are chosen based on the  
 141 monodispersed or bidispersed fluidized-bed configurations summarized in table I. The fluid  
 142 has density  $\rho_f = 998.21 \text{ kg m}^{-3}$  and kinematic viscosity  $\nu_f = 10^{-6} \text{ m}^2\text{s}^{-1}$ . The Cartesian  
 143 grid spacing is uniform and given by  $\Delta x = \Delta y = \Delta z = h$  such that

$$h = \frac{\max(d_{p,1}, d_{p,2}, d_{p,3})}{25.6} = \frac{\min(d_{p,1}, d_{p,2}, d_{p,3})}{18.3}, \quad (4)$$

144 which is sufficient to resolve the flow-particle interactions as demonstrated by various au-  
 145 thors [36–38] and demonstrated with our code in Ref [40]. The rectangular domain has a  
 146 square cross section  $L_x = L_y = 10d_{p,1}$  and a height  $L_z = 60d_{p,1}$ , giving a three-dimensional  
 147 grid with  $256 \times 256 \times 1536$  grid points. The time-step size  $\Delta t$  is calculated based on the advec-  
 148 tion and diffusion Courant number which are defined as  $C_{\text{adv}} = u_0 \Delta t / h$  and  $C_{\text{diff}} = \nu_f \Delta t / h^2$ ,  
 149 respectively, and we ensure that  $C_{\text{max}} = \max(C_{\text{adv}}, C_{\text{diff}}) = 0.25$  for the case with the largest  
 150 flow rate. In the simulations, the critical parameters are the particle diameter and the num-  
 152 ber of particles. Here, we choose the largest particle diameter  $d_{p,1} = 0.002 \text{ mm}$  to compare  
 153 the results to the monodispersed simulations of Ref [16], while  $d_{p,2}$  and  $d_{p,3}$  are calculated  
 154 based on assuming

$$d_{p,1} = 1.2d_{p,2} = 1.4d_{p,3}. \quad (5)$$

155 The choice of the diameter ratio is designed to minimize the computational cost (that scales  
 156 with the diameter ratio) while ensuring bidispersed behavior. For the monodispersed sim-  
 157 ulations, the number of particles  $N_{p,mono} = 2000$  is used to ensure a sufficient fluidized bed

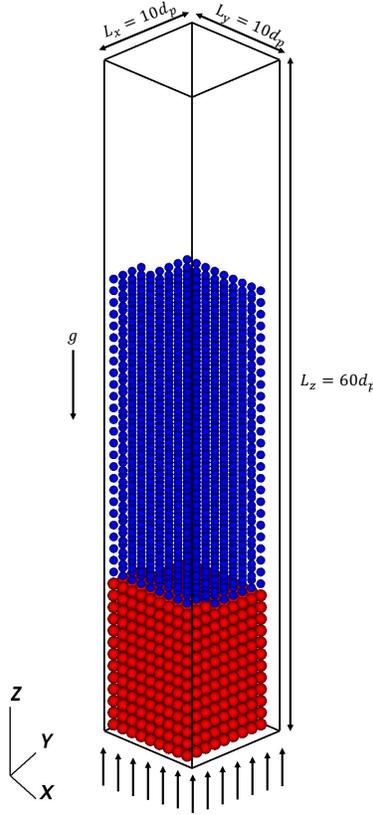


Figure 1. The three-dimensional computational domain, showing the bidispersed fluidized bed and the uniform inflow velocity profile. Particle positions are initialized with  $1d_{p,1}$  spacing for the simulation with  $Re_p = 40$  for FB-Bi-14.

158 height to obtain accurate statistics. For the bidispersed simulations, the number of particles  
 159  $N_{p,bi}$  varies from case to case and is defined as

$$N_{p,bi} = N_{p,d_{p,1}} + N_{p,d_{p,j}}, \quad (6)$$

160 where  $j = 2$  or  $3$ ,  $N_{p,d_{p,1}}$  and  $N_{p,d_{p,j}}$  are the number of particles with particle diameter  $d_{p,1}$   
 161 and  $d_{p,j}$ , respectively. To ensure a fluidized-bed height that is sufficiently high to obtain good  
 162 statistics, the bidispersed fluidized-bed height  $L_{bi}$  is kept the same as the monodispersed  
 163 fluidized-bed height. Defining the monodispersed bed height determined by Ref [16] as  
 164  $L_{mono,d_{p,1}}$ , we require

$$L_{bi} = L_{mono,d_{p,1}} = 2 \times L_{bi,d_{p,j}} = 2 \times L_{bi,d_{p,1}}. \quad (7)$$

	FB-Mono-12	FB-Bi-12		FB-Mono-14	FB-Bi-14		FB-Mono-10
$u_0$ (m s <sup>-1</sup> )	$N_{p,mono}$	$N_{p,d_{p,1}}$	$N_{p,d_{p,2}}$	$N_{p,mono}$	$N_{p,d_{p,1}}$	$N_{p,d_{p,3}}$	$N_{p,mono}$
0.010	2000	1000	1607	2000	1000	2376	2000
0.015	2000	1000	1565	2000	1000	2247	2000
0.020	2000	1000	1518	2000	1000	2106	2000
0.025	2000	1000	1465	2000	1000	1943	2000
0.030	2000	1000	1401	2000	1000	1749	2000
0.035	2000	1000	1321	2000	1000	2509	2000

Table I. Summary of number of particles used in the simulations.  $N_{p,mono} = 2000$  is used for all monodispersed cases.  $N_{p,d_{p,2}}$  and  $N_{p,d_{p,3}}$  are calculated using equation 8 with respect to the upflow velocity. Particles with diameter  $d_{p,1} = 2.0$  mm,  $d_{p,2} = d_{p,1}/1.2$  and  $d_{p,3} = d_{p,1}/1.4$  are used. Results from FB-Mono-10 were obtained from Ref [16]

165 Given  $L_{mono,d_{p,1}}$ ,  $N_{p,mono}$ ,  $d_{p,1}$  and  $d_{p,j}$ , if we assume the number of particles in the lower  
166 layer is  $N_{p,d_{p,1}} = 1000$ , then the number of particles in the upper layer is given by

$$N_{p,d_{p,j}} = \frac{L_x L_y L_{bi,d_{p,j}} \phi}{V_{p,d_{p,j}}}, \quad (8)$$

167 where  $V_{p,d_{p,j}} = \pi d_{p,j}^3 / 6$  is the volume of a particle with diameter  $d_{p,j}$  and  $\phi$  can be estimated  
168 with equations 1. Table I summarizes the number of particles used in each simulation such  
169 that the largest number of particles used is  $N_{p,bi} = 3376$ . This represents a good balance  
170 between ensuring a sufficient fluidized bed height while minimizing the number of particles,  
171 which significantly increase the computational cost.

172 In this work, in addition to the particle diameter ratio, the second parameter of interest  
173 is the particle Reynolds number. For a monodispersed fluidized bed, the particle Reynolds  
174 number is defined as  $Re_{p,i} = u_0 d_{p,i} / \nu_f$  where  $i = 1, 2$  or  $3$ . For a bidispersed fluidized-  
175 bed, two particle Reynolds numbers  $Re_{p,1}$  and  $Re_{p,j}$  are defined based on the two particle  
176 diameters  $d_{p,1}$  and  $d_{p,j}$  where  $j = 2$  or  $3$ . In order to maintain the same flow rate as the  
177 monodispersed simulations, we vary  $Re_{p,1}$ . In total, six simulations were conducted with  
178  $0.010 \text{ m s}^{-1} \leq u_0 \leq 0.035 \text{ m s}^{-1}$ , giving  $20 \leq Re_{p,1} \leq 70$  for each configuration summarized  
179 in table I, giving a total of 24 simulations. For all cases, the pressure is specified at the top  
180 boundary as  $p = 0$ , while a uniform inflow velocity of  $u_0$  is specified at the bottom boundary.

181 Simulations are initialized with a uniform array of particles and the flow is started from  
 182 rest. The upflow velocity leads to expansion of the bed and random motion of the particles  
 183 until statistical equilibrium is reached, at which time the dynamics are independent of the  
 184 initial particle distribution and the total average drag force is in balance with the submerged  
 185 weight of the particles. To understand the time evolution of particle variables and assess  
 186 statistical equilibrium, we define the naive ensemble-average operator in which data for all  
 187 particles are used as

$$\langle \{\cdot\} \rangle^* = \frac{1}{N_p} \sum_{n=1}^{N_p} \{\cdot\}_n, \quad (9)$$

188 where  $N_p$  is the total number of particles in the simulation, and we monitor the ensemble-  
 189 average vertical particle velocity  $\langle w_p \rangle^*$ . Here, we define a turnover time  $\tau_T = d_{p,\max}/u_0$  and  
 190 statistical equilibrium is achieved after  $30\tau_T$  that is indicated by  $\langle w_p \rangle^*$  fluctuating about zero.  
 191 Figure 2 shows  $\langle w_p \rangle^*$  as a function of maximum turnover time  $\tau_T$  for different configurations  
 192 with the same  $Re_{p,1} = 40$ . The  $\langle w_p \rangle^*$  of both monodispersed and bidispersed fluidized bed  
 193 converges to zero, indicating statistical equilibrium. Here, we define the time-averaging  
 194 operator

$$\overline{\{\cdot\}} = \frac{1}{t_{\max} - t_i} \int_{t_i}^{t_{\max}} \{\cdot\} dt, \quad (10)$$

195 where  $t_i = t_0 + 30\tau_T$  and  $t_0$  is the spin-up time needed for the flow to reach statistical  
 196 equilibrium.

### 197 III. RESULTS

#### 198 A. Distribution of volume fraction in the bidispersed fluidized bed

199 Many researchers have reported that segregated bidispersed fluidized beds consist of  
 200 three different regions (lower, transition and upper layer) [13, 19, 20]. The volume fraction  
 201 of the lower and upper layers can be approximated with the corresponding values for a  
 202 monodispersed fluidized bed. To validate the volume fraction distribution of the segregated  
 203 bidispersed fluidized bed, we compute the instantaneous Eulerian volume fraction  $\phi(\mathbf{x}, t)$   
 204 following the procedure in Ref [16]. We compute the volume fraction as a function of

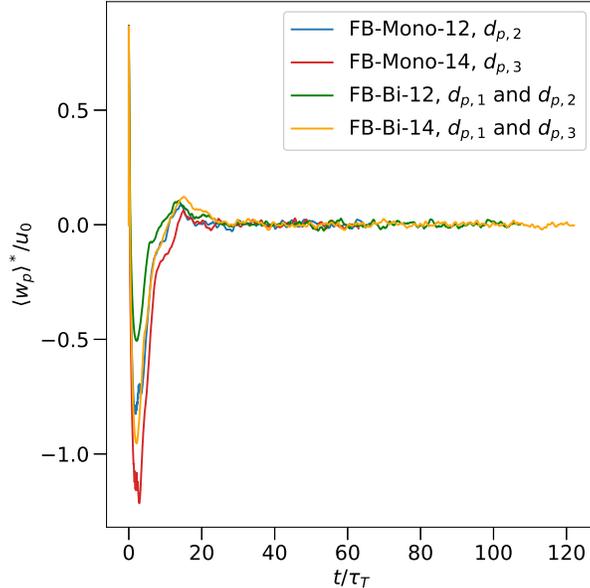


Figure 2. Time series of the ensemble settling velocity  $\langle w_p \rangle^*$  normalized by  $u_0$  for  $Re_{p,1} = 40$  for all cases simulated except FB-Mono-10.

vertical position by applying the Eulerian horizontal-averaging operator

$$\langle \{ \cdot \} \rangle_{xy}^* = \frac{1}{N_x N_y} \sum_{i,j=1}^{N_x N_y} \{ \cdot \}_{ij}. \quad (11)$$

Figure 3 shows  $\langle \bar{\phi} \rangle_{xy}^*(z)$  as a function of  $z/d_{p,1}$  for different  $Re_{p,1}$ . Qualitatively, the  $\langle \bar{\phi} \rangle_{xy}^*(z)$  of FB-Mono-12 and FB-Mono-14 approximately match the corresponding upper layers of FB-Bi-12 and FB-Bi-14. For quantitative comparison, we compute the vertically-averaged  $\langle \bar{\phi} \rangle_{xy}^*(z)$  volume fraction  $\langle \langle \bar{\phi} \rangle_{xy}^* \rangle_z$  by excluding the boundaries. To do so, we define a modified Eulerian vertical-averaging operator

$$\langle \{ \cdot \} \rangle_z = \frac{1}{N_z^*} \sum_{k=z_b/h}^{z_t/h} \{ \cdot \}_k, \quad (12)$$

where  $z_b$  and  $z_t$  are the bottom and top of the homogeneous fluidized-bed layers, respectively, and  $N_z^* = (z_t - z_b)/h$  is the number of grid points in the  $z$ -direction bounded by  $z_b$  and  $z_t$ . ‘‘Homogeneous’’ refers to the region of the fluidized-bed that is not affected by the boundaries. In what follows,  $\langle \langle \bar{\phi} \rangle_{xy}^* \rangle_z \equiv \langle \bar{\phi} \rangle$  will be assumed unless otherwise indicated.

For FB-Mono-12 and FB-Mono-14, we define  $z_b$  and  $z_t$  following the procedure in Ref [16] by excluding the values near the top and bottom of the fluidized bed. For FB-Bi-12 and FB-Bi-14, the fluidized beds consist of three regions with a total of four boundaries (two for each

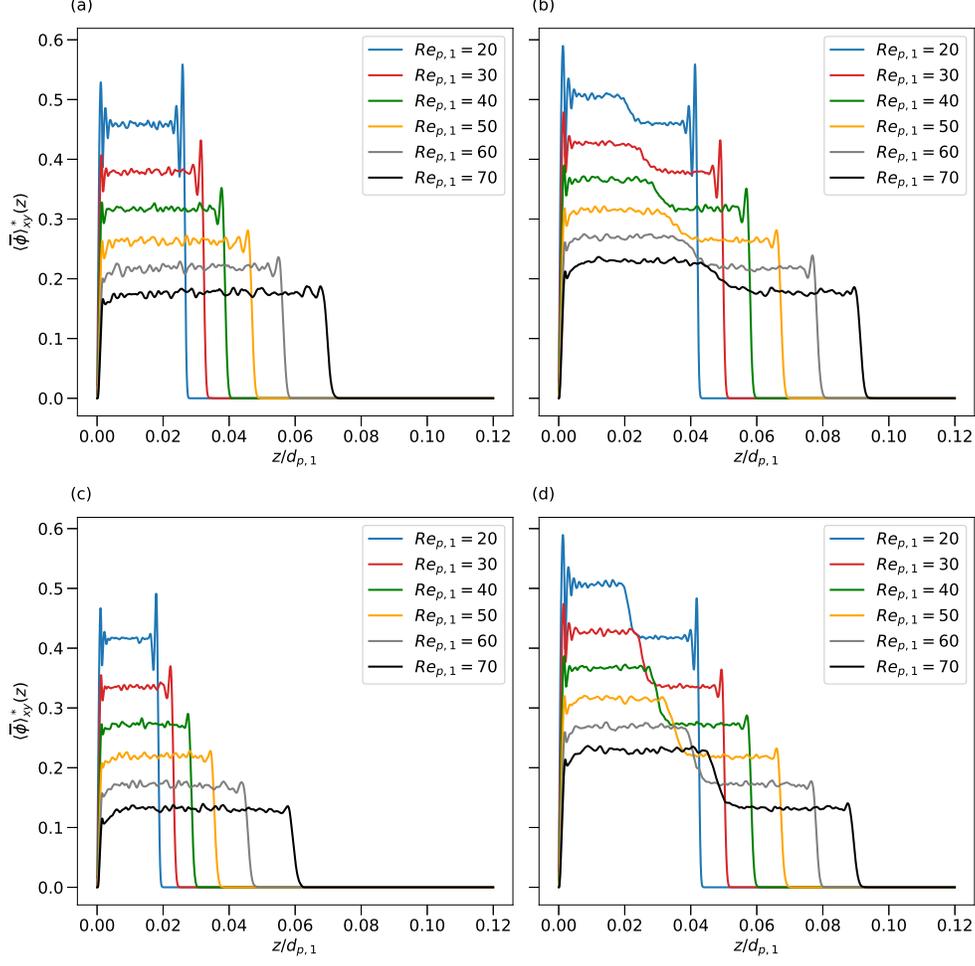


Figure 3. Horizontally- and time-averaged volume fraction  $\langle \bar{\phi} \rangle_{xy}^*$  as a function of normalized vertical position  $z/d_{p,1}$  for different particle Reynolds numbers  $Re_{p,1}$ . (a) FB-Mono-12. (b) FB-Bi-12. (c) FB-Mono-14. (d) FB-Bi-14.

218 segregated layer). In the transition region, the lower layer  $\langle \bar{\phi} \rangle$  decreases monotonically from  
 219 the lower to the upper layer  $\langle \bar{\phi} \rangle$  as shown in figure 3(b) and (d). To define the boundaries of  
 220 each region, we construct probability density functions (PDF) based on the particle vertical  
 221 position with respect to particle diameter as

$$\mathbf{P}_\alpha(z) = \frac{1}{N_t} \sum_{t=1}^{N_t} \sum_{i=1}^{N_p} [\mathbf{1}_{z_l < z_{p,i}^t < z_u} (z_{p,i}^t)] [\mathbf{1}_{d_{p,i} = d_{p,j}} (d_{p,i})], \quad (13)$$

222 where  $z_{p,i}$  is the vertical position of the  $i$ th particle,  $z_l$  and  $z_u$  denote the lower and upper  
 223 edges of the equally-spaced bins having a width of  $0.5d_{p,1}$ ,  $N_p$  is the total number of particles

224 in the fluidized bed,  $\mathbf{1}_{z_l < z_{p,i}^t < z_u}(z_{p,i}^t)$  is the indicator function

$$\mathbf{1}_{z_l < z_{p,i}^t < z_u}(z_{p,i}^t) \begin{cases} 1 & z_l < z_{p,i}^t < z_u, \\ 0 & \text{otherwise,} \end{cases} \quad (14)$$

225 that determines whether particle  $i$  is located in the bin bounded by  $z_l$  and  $z_u$ , and  
 226  $\mathbf{1}_{d_{p,i}=d_{p,j}}(d_{p,i})$  is the indicator function

$$\mathbf{1}_{d_{p,i}=d_{p,j}}(d_{p,i}) \begin{cases} 1 & d_{p,i} = d_{p,j}, \\ 0 & \text{otherwise,} \end{cases} \quad (15)$$

227 to include particles with diameter  $d_{p,j}$ . Figure 4 shows the PDF of the particle vertical  
 228 position for case FB-Bi-12 with  $Re_{p,1} = 40$ . We define the top of the lower layer as

$$z_{t,lower} = \arg \min \left( \mathbf{P}_{d_{p,1}}(z) / \mathbf{P}_{d_{p,j}}(z) - \zeta_{thresh} \right) \quad (16)$$

229 and the bottom of the upper layer as

$$z_{b,upper} = \arg \min \left( \mathbf{P}_{d_{p,j}}(z) / \mathbf{P}_{d_{p,1}}(z) - \zeta_{thresh} \right), \quad (17)$$

230 where  $\zeta_{thresh} = 100$  is an arbitrary threshold ratio to be set apriori. Large  $\zeta_{thresh}$  will result  
 231 in a more monodispersed-like segregated layer that leads to a larger transition region and  
 232 smaller segregated region. Table II summarizes  $z_{t,lower}$  and  $z_{b,upper}$  for FB-Bi-12 and FB-Bi-  
 233 14. With the boundaries defined in this way, we can compute  $\langle \bar{\phi} \rangle$  for each segregated layer  
 234 in cases FB-Bi-12 and FB-Bi-14.

235 Figure 5 shows the fit of  $1 - \langle \bar{\phi} \rangle$  as a function of  $Re_{p,1}$  for different regions. Each  
 236 bidispersed fluidized bed can fit two different lines for the upper and lower layers, resulting  
 237 in a total of 7 fitted lines (only three can be seen in the figure due to overlap). Overall, FB-  
 238 Mono-12 [16] overlaps with the lower layer of FB-Bi-12 and FB-Bi-14 while FB-Mono-12 and  
 239 FB-Mono-14 overlaps with the corresponding upper layer of FB-Bi-12 and FB-Bi-14. This  
 240 shows that the volume fractions in the different layers of a segregated bidispersed fluidized-  
 241 bed can be approximated accurately with those of a monodispersed fluidized bed. Table III  
 242 summarizes the fitted  $n$  that are closer to the predicted  $n$  by Ref [21] and fitted  $k$  that is in  
 243 the same range as reported by various authors [8, 10]. Overall,  $n$  and  $k$  obtained from the  
 244 monodispersed fluidized bed and corresponding layers in the bidispersed fluidized bed are  
 245 indistinguishable, further demonstrating the accuracy of approximating the volume fraction  
 246 in each segregated layer as that in a monodispersed layer without boundary effects.

$u_0$ (m s <sup>-1</sup> )	FB-Bi-12		FB-Bi-14	
	$z_{t,lower}/d_{p,1}$	$z_{b,upper}/d_{p,1}$	$z_{t,lower}/d_{p,1}$	$z_{b,upper}/d_{p,1}$
0.010	8.44	12.7	9.35	25.9
0.015	8.74	16.3	11.2	22.0
0.020	11.5	17.5	12.7	19.3
0.025	13.3	22.0	14.5	16.6
0.030	15.4	23.8	17.2	14.2
0.035	18.4	27.7	20.5	11.8

Table II. Summary of the top boundary of lower layer  $z_{t,lower}$  and the bottom boundary of upper layer  $z_{b,upper}$  in the bidispersed fluidized bed. The transition region is defined as the difference between  $z_{t,lower}$  and  $z_{b,upper}$ .

Parameters	$d_{p,1}$			$d_{p,2}$		$d_{p,3}$	
	FB-Mono-10	FB-Bi-12	FB-Bi-14	FB-Mono-12	FB-Bi-12	FB-Mono-14	FB-Bi-14
$n$	2.81	2.83	2.81	3.00	2.99	3.15	3.14
$k$	0.71	0.72	0.72	0.74	0.74	0.76	0.76
$n_{ga}$	2.89			2.95		3.02	
$n_{zaki}$	2.61			2.71		2.80	

Table III. Summary of fitted  $n$  and  $k$  with respect to each monodispersed fluidized bed and segregated layers in the bidispersed fluidized-bed. All coefficients of determination  $R^2$  for these fits are 1.  $n_{zaki}$  and  $n_{ga}$  are calculated using Refs [7] and [21] respectively.

## 248 B. Kinematic wave speed

249 In the previous section, the fluidized bed is characterized by the volume fraction and  
250 particle Reynolds number. However, the volume fraction fluctuates about a mean value  
251 which exhibits alternating regions of low and high volume fractions, resulting in waves [41–  
252 43]. Comparison between the wave speed of segregated bidispersed and monodispersed  
253 fluidized beds will further validate the assumption of approximating segregated bidispersed  
254 fluidized beds as a superposition of two monodispersed fluidized beds.

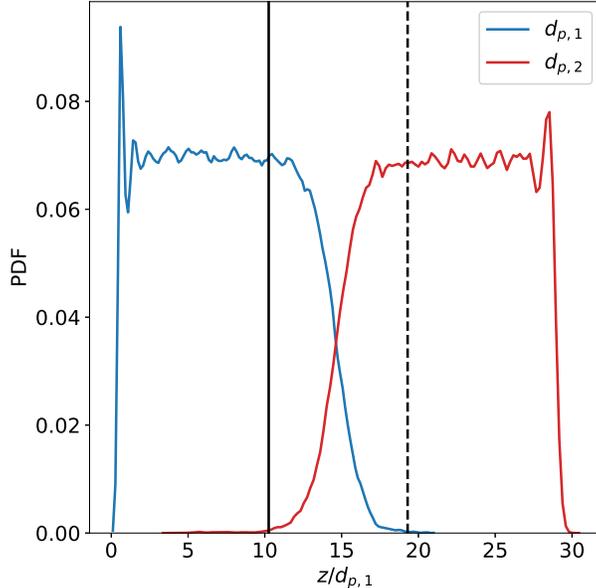


Figure 4. Probability density function (PDF) of particle vertical positions for the two particle diameters in the case with  $Re_{p,1} = 40$  for FB-Bi-12.

255 Based on the classification of fluidization by Refs [41, 42], the cases simulated in this work  
 256 are classified as unstable fluidization that is characterized by persistent particle velocity fluc-  
 257 tuations. Figure S1(a) shows a typical volume fraction fluctuation  $\phi' = \langle \bar{\phi} \rangle_{xy} - \langle \bar{\phi} \rangle$  plot in  
 258 two-dimensional space-time. Qualitatively, propagating waves are indicated by regions of  
 259 porosity that are periodic in space and time. However, due to the random noise, extract-  
 260 ing wave speeds from Figure S1(a) is difficult. In the Appendix B, wave speeds estimated  
 261 with three different approaches are compared, namely 1) naive, 2) two-dimensional auto-  
 262 correlation and 3) the dispersion relationship. The results indicate that the autocorrelation  
 263 approach is the most accurate and is adopted in this paper.

264 Ref [44] relates volume fraction to wave speed with

$$c = kn\phi(1 - \phi)^{n-1}w_{ref}, \quad (18)$$

265 where  $c$  is wave speed and other variables are consistent with equation 1. Figure 6(a),  
 266 (b) and (c) show the wave speeds computed with different particle diameters using the  
 267 autocorrelation approach and model (equation 18). Overall, the computed wave speeds are  
 268 very similar to the wave speeds computed with the model. Interestingly, the wave speed  
 269 in the different layers in the FB-Bi-12 and FB-Bi-14 cases agree with the corresponding

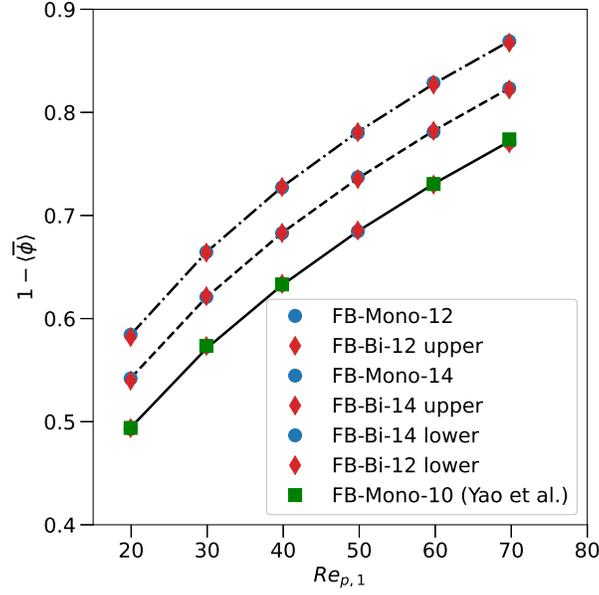


Figure 5. Porosity  $1 - \langle \bar{\phi} \rangle$  as a function of  $Re_{p,1}$  for the simulated monodispersed and bidispersed cases. The lines were constructed based on fitting to the power law equation (1).

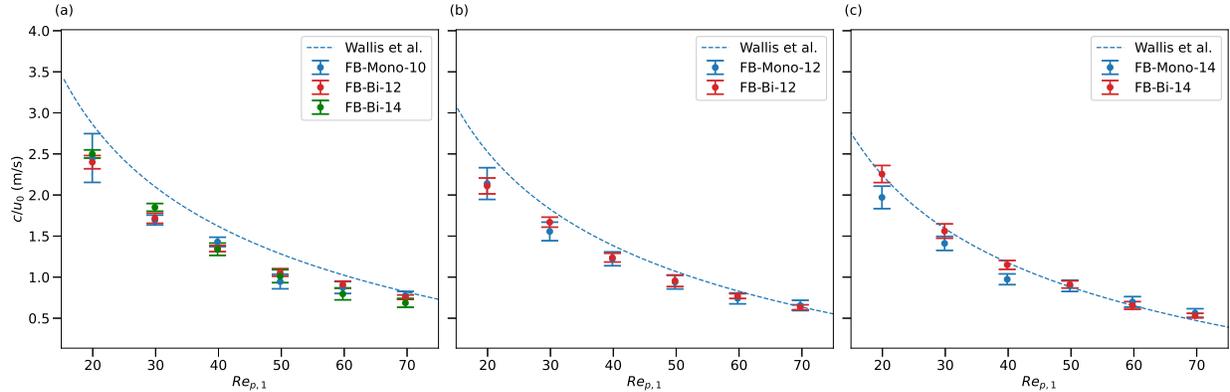


Figure 6. Wave speed based on the autocorrelation as a function of  $Re_{p,1}$  for (a)  $d_{p,1}$ , (b)  $d_{p,2}$  and (c)  $d_{p,3}$ .

270 monodispersed fluidized-bed cases even though the boundary conditions on each segregated  
 271 layer in cases FB-Bi-12 and FB-Bi-14 are different. For illustration, case FB-Mono-12 is  
 272 prescribed with a uniform inflow profile while the upper layer of case FB-Bi-12 is subjected  
 273 to the non-uniform flow at the transition region. This shows that the wave speed is controlled  
 274 by the particle properties and local porosity rather than the inflow.

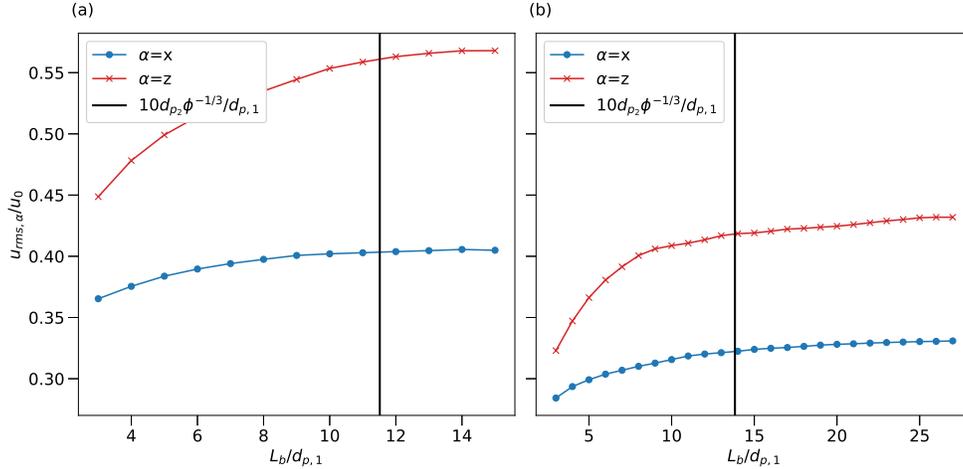


Figure 7. Particle velocity fluctuations as a function of the fluidized-bed height  $L_b$  for case FB-Mono-12 with (a)  $Re_{p,1} = 30$  and (b)  $Re_{p,1} = 60$ .

### 275 C. Velocity fluctuations

276 To understand the effect of particle velocity fluctuations in a segregated bidispersed  
 277 fluidized bed, we compute the root-mean-square velocity

$$u_{rms,\alpha} = \sqrt{\langle u'_\alpha u'_\alpha \rangle}, \quad (19)$$

278 where  $u'_\alpha = u_\alpha - \langle \bar{u} \rangle_\alpha$  is the particle velocity fluctuation and  $\alpha = x, y$  or  $z$ . In a suspended  
 279 particle system, Ref [23] discovered that particle velocity fluctuations depend on the domain  
 280 size for  $\min(L_x, L_y, L_b) < 10d_{p,\max}\phi^{-1/3}$  and otherwise scale as  $2w_t\phi^{1/3}$ . By simulating Stokes  
 281 sedimentation, Ref [45] demonstrated that the dependence on the domain size exists with  
 282 periodic horizontal and non-periodic z-direction boundaries. Figure 7 shows the particle  
 283 velocity fluctuations  $u'_\alpha$  for case FB-Mono-12 as a function of the normalized bed height  
 284 for  $Re_{p,1} = 30$  and 60. Particle velocity fluctuations initially increase at a higher rate as  
 285  $L_b$  increases. When  $L_b$  is sufficiently large (above the black line), the increase in particle  
 286 velocity fluctuations is less significant. Since the heights of the segregated layers in cases FB-  
 287 Bi-12 and FB-Bi-14 are less than the critical height  $10d_{p,\max}\phi^{-1/3}$ , they differ from the height  
 288 of the corresponding monodispersed fluidized bed. Therefore, particle velocity fluctuations  
 289 in the monodispersed fluidized bed are calculated with particles located below the desired  
 290 height to eliminate the effect of  $L_b$  on particle velocity fluctuations. Figure 8(a) and (b)  
 291 show the normalized particle velocity fluctuations as a function of  $Re_{p,1}$ . With the same  
 292

293 bed height, the normalized particle velocity fluctuations for cases FB-Mono-10, FB-Mono-12  
 294 and FB-Mono-14 are nearly identical to those of the corresponding layers in the bidispersed  
 295 fluidized-bed cases. At low Reynolds numbers ( $Re_{p,1} \leq 40$ ), the upper segregated layer has  
 296 the largest normalized particle velocity fluctuations, followed by the transition and lower  
 297 layers. As the Reynolds number increases, particle velocity fluctuations in the transition  
 298 layer become greater than those of the upper segregated layer. For  $Re_{p,d_p,1} > 60$ , the particle  
 299 velocity fluctuations in the transition layer are the highest, followed by the lower and upper  
 300 layers. The trends in figure 8 indicate that the normalized particle velocity fluctuations of  
 301 the lower layers will eventually become the largest. This observation is due to the different  
 302 particle properties and volume fraction operating at a common  $Re_{p,1}$ , resulting in different  
 303 porosity due to different particle properties. To eliminate this effect, figure 8(c) and (d)  
 304 show the normalized particle velocity fluctuations as a function of the porosity  $1 - \langle \bar{\phi} \rangle$ . The  
 305 normalized particle velocity fluctuations are expected to be zero for both a single particle  
 306 ( $\phi \approx 0$ ) and a packed bed ( $\phi \approx 0.6$ ), resulting in a maximum value at an intermediate  
 307 porosity. The upper segregated layer has consistently higher normalized particle velocity  
 308 fluctuations than the lower segregated layer at the same porosity due to the decreasing  
 309 particle cluster lifespan with increasing Archimedes number [40], which is defined as

$$Ar = \frac{g(s-1)d_{p,i}^3}{\nu_f^2}, \quad (20)$$

310 where  $i = 1, 2$  or  $3$ ,  $d_{p,i}$  is the diameter of particle  $i$ ,  $g$  is the gravitational acceleration  
 311 and  $s = \rho_p/\rho_f$  is the particle-fluid density ratio. According to Ref [40], particles are more  
 312 likely to form long-lived clusters for particles with lower  $Ar$  due to ineffective collisions that  
 313 are unlikely to break particle clusters formed by wake entrainment. With more long-lived  
 314 clusters, particles experience appreciable acceleration as a cluster resulting in more signifi-  
 315 cant normalized particle velocity fluctuations. Despite higher particle velocity fluctuations  
 316 for lower  $Ar$ , we would like to point out that figure 8 resembles more realistic fluidized-bed  
 317 operation. With a common upflow velocity, particles with different  $Ar$  cannot be operated  
 318 at the same porosity. To optimize reactor mixing that is likely to coincide with the peak in  
 319 particle velocity fluctuations [16], both layers must be considered separately.

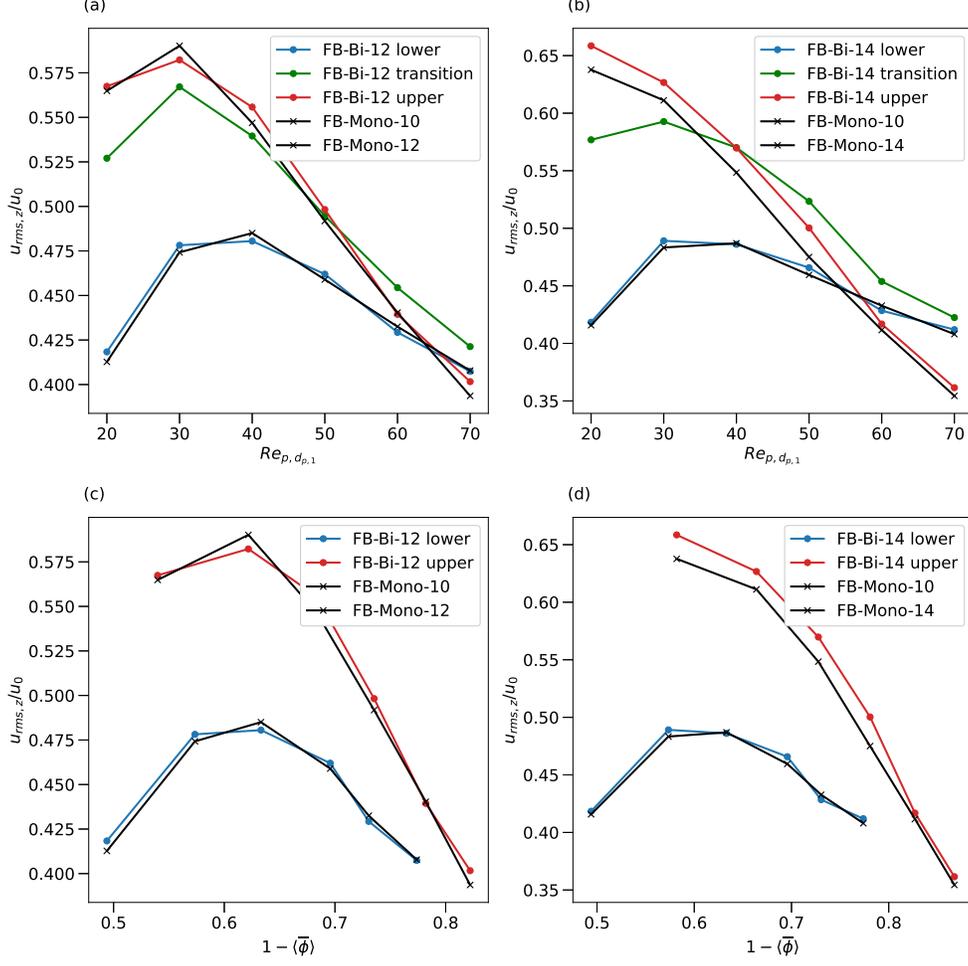


Figure 8. Normalized particle velocity fluctuations as a function of  $Re_{p,1}$  for (a)  $d_{p,1}/d_{p,2} = 1.2$  and  $d_{p,1}/d_{p,3} = 1.4$  and  $1 - \langle \bar{\phi} \rangle$  for (c)  $d_{p,1}/d_{p,2} = 1.2$  and (d)  $d_{p,1}/d_{p,3} = 1.4$ .

#### 320 D. Auto-correlation and self-diffusivity

In this section, we compute the integral timescale and self-diffusivity for different regions in the bidispersed fluidized bed and compare them to the corresponding monodispersed fluidized bed. As defined by Refs [9, 10, 46], the autocorrelation function is given by

$$R_{\alpha\alpha}(\tau) = \frac{\langle u'_\alpha(t_0)u'_\alpha(t_0 + \tau) \rangle}{\langle (u'_\alpha(t_0))^2 \rangle}, \quad (21)$$

321 where  $\tau$  is the given time lag and  $\alpha = x, y$  or  $z$ . Following the procedure to quantify the  
 322 errors in computing the integral timescale due to finite simulation time in [16], we compute

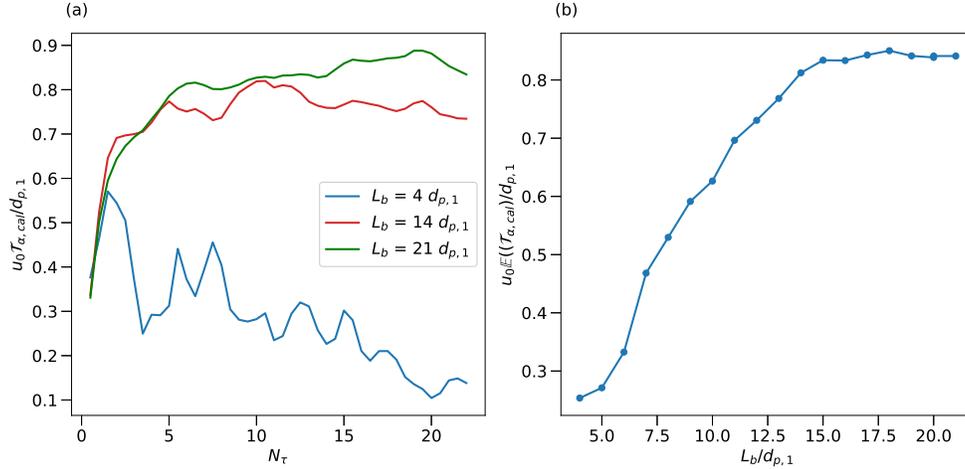


Figure 9. Calculated integral timescale as a function of (a) simulated duration  $N_\tau$  and (b) fluidized-bed height  $L_b/d_{p,1}$  for case FB-Mono-12 with  $Re_{p,1} = 40$ .

323 the approximate integral timescale as

$$\mathbb{E}(\mathcal{T}_{\alpha,cal}) = \frac{1}{N_\tau - N_{\tau,thresh}} \sum_{i=N_{\tau,thresh}}^{N_\tau} \mathcal{T}_{\alpha,cal}^i, \quad (22)$$

$$\text{STD.}(\mathcal{T}_{\alpha,cal}) = \sqrt{\mathbb{E}(\mathcal{T}_{\alpha,cal}^2) - \mathbb{E}(\mathcal{T}_{\alpha,cal})^2}, \quad (23)$$

where  $N_\tau = t/\tau_T$  and  $N_{\tau,thresh}$  is the threshold time needed to reach statistical equilibrium, and the calculated integral timescale with  $N_\tau$  is defined as

$$\mathcal{T}_{\alpha,cal}^{N_\tau} = \int_0^{t_f} R_{\alpha\alpha}(\tau) d\tau. \quad (24)$$

324 Figure 9(a) shows the effects of  $N_\tau$  on the computed integral timescale for the entire flu-  
 325 idized bed. For each respective fluidized-bed height,  $\mathcal{T}_{\alpha,cal}$  initially increases as  $N_\tau$  increases  
 326 and fluctuates about a mean value after  $N_\tau \approx 10$ , demonstrating that the computed in-  
 327 tegral timescale has converged in time. The fluctuations are likely due to the presence  
 328 of waves in the fluidized bed which produce alternating positive and negative autocorrela-  
 329 tions [9, 16]. We also compute  $\mathcal{T}_{\alpha,cal}$  as a function of  $L_b$  by considering particles that are  
 331 located in the desired range of the fluidized bed for 99% of the simulated duration. As  
 332 shown in figure 9(b),  $\mathbb{E}(\mathcal{T}_{\alpha,cal})$  converges as  $L_b/d_{p,1}$  increases. After  $L_b > L_{b,crit}$ , the integral  
 333 timescale is independent of the fluidized-bed height. Since  $L_b < L_{b,crit}$  for cases FB-Bi-12  
 334 and FB-Bi-14, we adopt a similar approach as Section III C by computing a reduced  $L_b$

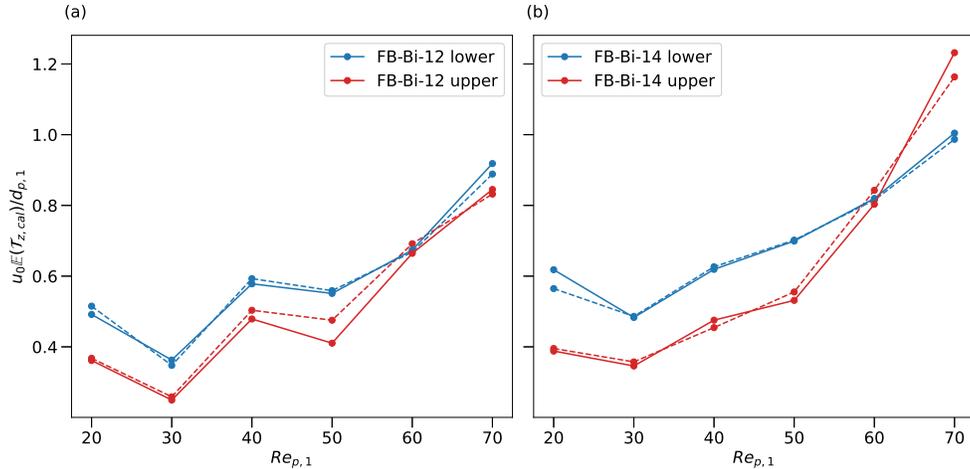


Figure 10. Expected integral timescale as a function of  $Re_{p,1}$  for (a)  $d_{p,1}/d_{p,2} = 1.2$  and  $d_{p,1}/d_{p,3} = 1.4$ .

335 for the monodispersed simulations (cases FB-Mono-12 and FB-Mono-14). As shown in fig-  
 336 ure 10, the integral timescale for the bidispersed cases are of the same order of magnitude  
 337 as the truncated monodispersed integral timescales. The non-monotonic behavior is due to  
 338 the error associated with  $L_b < L_{b,crit}$ . Since the main focus is not on the absolute magni-  
 339 tude but on the relative magnitude between the monodispersed and bidispersed fluidized  
 340 beds, approximating the segregated layers in bidispersed fluidized beds with corresponding  
 342 monodispersed fluidized beds is still valid.

Following Refs [9, 46], the self-diffusivity is defined as

$$\mathcal{D}_{\alpha,cal}^{N\tau} = \int_0^{t_f} R_{\alpha\alpha}(\tau) \langle (u'_\alpha(t_0))^2 \rangle d\tau. \quad (25)$$

343 Similar to the integral timescale, the self-diffusivity initially depends on  $L_b$  until  $L_b > L_{b,crit}$   
 344 (not shown). Therefore, to compare to the bidispersed cases, we compute the truncated  
 345 self-diffusivity for the monodispersed cases. Figure 11 shows that the self-diffusivity of the  
 346 bidispersed cases is comparable to the self-diffusivity of the monodispersed cases, indicating  
 347 the validity of characterizing bidispersed fluidized beds using properties of the corresponding  
 349 monodispersed layers.

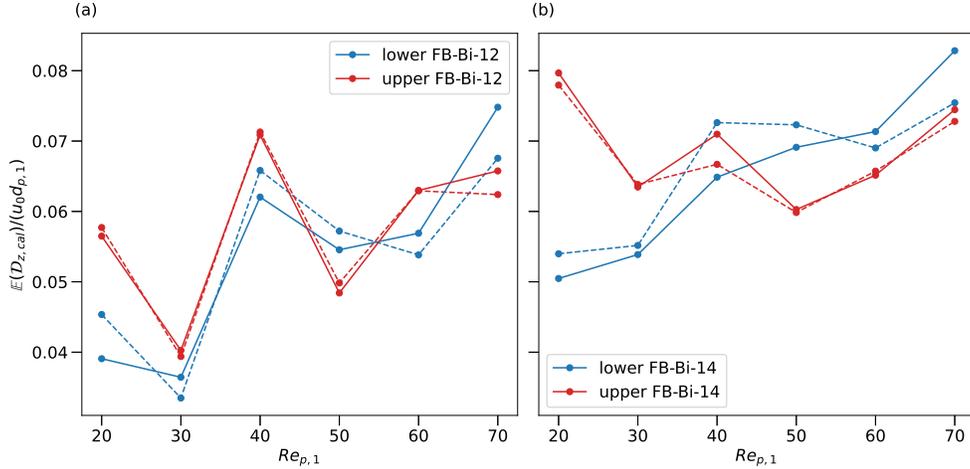


Figure 11. Self-diffusivity as a function of  $Re_{p,1}$  for (a)  $d_{p,1}/d_{p,2} = 1.2$  and  $d_{p,1}/d_{p,3} = 1.4$ .

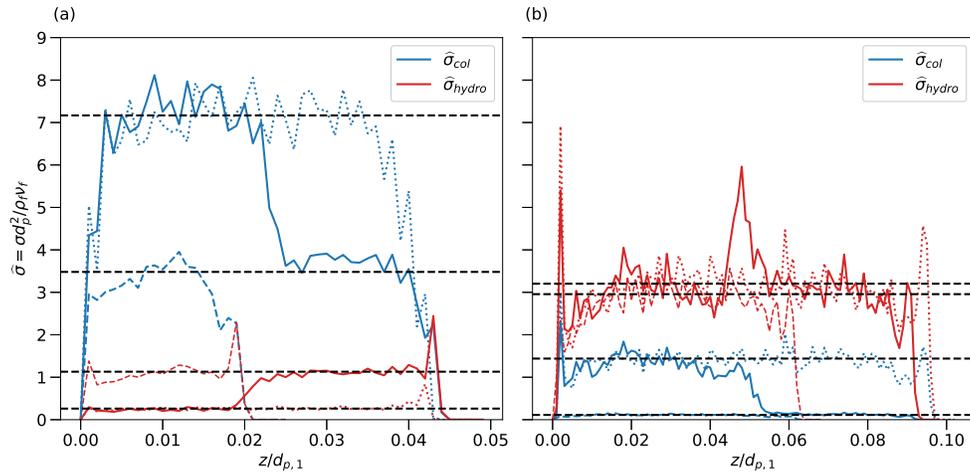


Figure 12. Normal contact stress and hydrodynamic stress as a function of vertical position for  $d_{p,1}/d_{p,3} = 1.40$  and with (a)  $Re_{1,p} = 20$  and (b)  $Re_{1,p} = 70$ . Solid lines correspond to bidispersed fluidized-bed layers and dotted lines correspond to monodispersed layers.

### 350 E. Particle-particle and fluid-particle interactions

351 Ref [16] showed that the dominant mechanism inducing particle velocity fluctuations  
 352 shifts from collisions to hydrodynamic forces as the particle Reynolds number increases.  
 353 Following the approach outlined in Ref [16], we compute the normal contact stress  $\sigma_{col}$ , nor-  
 354 mal lubrication stress  $\sigma_{lub}$  and hydrodynamic stresses  $\sigma_{hydro}$  in the bidispersed fluidized bed.  
 355 Figure 12 shows the magnitude of each stress as a function of vertical position for  $Re_{p,1} = 20$   
 356

357 and 70 for cases FB-Bi-14, FB-Mono-10 and FB-Mono-14. For the range of Reynolds num-  
358 ber simulated, lubrication stresses are negligible (not shown). At low Reynolds numbers for  
359 the bidispersed simulation, the normal contact stress smoothly transitions from a high value  
360 in the lower layer to a lower value in the upper layer because of the reduced likelihood of  
361 collisions in the higher-porosity upper layer. The collision stresses in the upper and lower  
362 layers are roughly equal to the stresses in the corresponding monodispersed cases, and the  
363 hydrodynamic stresses are negligible due to the low upflow velocity. At high Reynolds num-  
364 bers, similar trends are observed in which the normal contact stress decreases monotonically  
365 to zero moving from the lower to the upper layers. Unlike the collision stress, however, the  
366 hydrodynamic stress peaks in the transition region rather than monotonically decreasing  
367 from the lower to the upper layers. In the transition region at higher Reynolds numbers,  
368 more vigorous velocity fluctuations are induced when large particles coexist with small par-  
369 ticles because small particles are strongly affected by the wakes of the large particles, thus  
370 leading to a peak in the hydrodynamic stress in the transition region.

372 Figure 13 shows that the collision stresses decrease monotonically from the lower to  
373 the upper layers for all cases, indicating a strong dependence of the collision stress on the  
374 Archimedes number in each layer. In addition, the collision stresses in the lower and up-  
375 per layers of the bidispersed fluidized-bed match those of the corresponding monodispersed  
376 fluidized-bed. This shows that collision stresses in the bidispersed fluidized bed can be  
377 approximated as those in the monodispersed fluidized bed. However, because the hydro-  
378 dynamic stress is a weaker function of the Archimedes number in each layer but a strong  
379 function of particle Reynolds number, the hydrodynamic stresses in the transition region  
380 are greater than those in the lower and upper layers in the bidispersed fluidized bed. Nev-  
381 ertheless, this analysis demonstrates that both collision and hydrodynamic stresses in the  
382 lower and upper layers of a segregated bidispersed fluidized bed can be approximated by the  
383 corresponding values in a monodispersed fluidized-bed.

#### 384 IV. CONCLUSION

385 We utilized PRS to compare the effects of the particle Reynolds number and bidispersity  
386 on both macroscopic and microscopic behavior of a fluidized bed in a three-dimensional  
387 domain. The particle Reynolds number was varied by varying the flow rate suspending

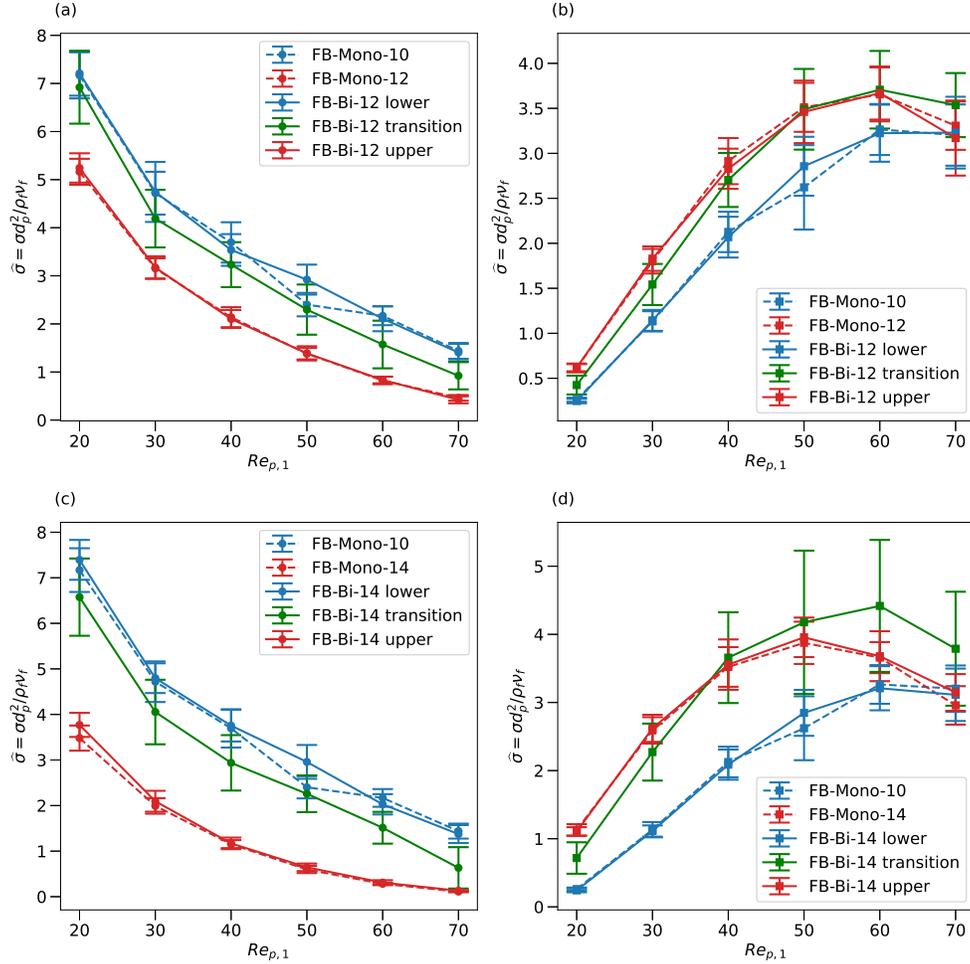


Figure 13. The computed normal contact stress and hydrodynamic stress as a function Reynolds number  $Re_{p,1}$ . (a) normal contact stress for  $d_{p,1}/d_{p,2} = 1.2$ . (b) hydrodynamic stress for  $d_{p,1}/d_{p,2} = 1.2$ . (c) normal contact stress for  $d_{p,1}/d_{p,3} = 1.4$ . (d) hydrodynamic stress for  $d_{p,1}/d_{p,3} = 1.4$ .

388 particles in the axial direction. Analysis of various statistics provided detailed comparison  
 389 between monodispersed and bidispersed fluidized beds. We have validated the assumption in  
 390 approximating the volume fraction of segregated bidispersed fluidized bed with the volume  
 391 fraction of two corresponding monodispersed fluidized beds. Fitting the porosity  $1 - \langle \bar{\phi} \rangle$  to  
 392  $Re_{p,1}$  further confirms that each segregated layer in the bidispersed fluidized-bed behaves  
 393 like a monodispersed fluidized-bed and can be calculated using a power-law relationship.  
 394 To understand the effects of wave speed in the monodispersed and bidispersed fluidized  
 395 beds, we filter out random noise in the volume fraction fluctuation using a low-pass filter  
 396 and approximate the wave speed using three different approaches. As compared to the

397 corresponding monodispersed fluidized bed with uniform inflow conditions, the upper layer  
398 of a bidispersed fluidized bed has an equivalent non-uniform flow conditions due to the  
399 transition layer. For a inflow-dominated wave speed, the wave speed of the upper layer of the  
400 bidispersed fluidized bed is expected to differ from that of the corresponding monodispersed  
401 fluidized bed. Results show that the non-uniform flow that fluidizes the upper layer of a  
402 bidispersed fluidized bed does not result in a different wave speed. This reveals that the  
403 wave speed is instead controlled by the local porosity rather than the inflow conditions. As  
404 the fluid flows through the voids between particles, inflow conditions are no longer important  
405 in determining the wave speed of the volume fraction fluctuations. Within the computed  
406 uncertainty bounds, the wave speed in each layer in the bidispersed fluidized bed agrees  
407 with the wave speed in the corresponding monodispersed fluidized bed.

408 Examination of particle velocity fluctuations shows that they are a strong function of  
409 the fluidized-bed height until the fluidized-bed height is greater than the critical bed height  
410  $10d_p\phi^{-1/3}$ . Due to limitations related to computational cost, the heights of the lower and  
411 upper layers of the bidispersed fluidized bed are less than the critical bed height. Therefore,  
412 we compute the particle velocity fluctuations of the monodispersed fluidized beds with bed  
413 heights equivalent to the corresponding upper and lower layers of the bidispersed fluidized  
414 beds. By computing particle velocity fluctuations in the monodispersed fluidized bed with  
415 equivalent heights, we have shown that the particle velocity fluctuations in the bidispersed  
416 fluidized bed match those of the corresponding monodispersed fluidized bed. Similarly, the  
417 convergence of the integral timescales and self-diffusivity are affected by both the fluidized-  
418 bed height and simulated duration until a critical bed height and sufficient long simulated  
419 duration are attained. Results show that the simulated duration of both the monodis-  
420 persed and bidispersed fluidized beds is sufficient. However, the bed heights of the upper  
421 and lower layers are insufficient to obtain converged statistics. In order to compare with  
422 the corresponding monodispersed fluidized beds, we compute the integral timescale of the  
423 monodispersed fluidized beds with an equivalent bed height to the corresponding upper and  
424 lower layers in the bidispersed fluidized beds. Using this approach, we confirmed that both  
425 self-diffusivity and integral timescales in the bidispersed fluidized-bed can be approximated  
426 by their corresponding values for a monodispersed fluidized bed.

427 By quantifying the lubrication, collision and hydrodynamic stresses, we showed that colli-  
428 sion stresses are a strong function of both Archimedes number and particle Reynolds number,

429 while hydrodynamic stresses depend more strongly on the particle Reynolds number. Fur-  
430 thermore, the collision stress in the bidispersed fluidized bed decreases monotonically from  
431 the lower to the upper layer while the hydrodynamic stress has a peak in the transition  
432 region at high Reynolds number. This points out the need to develop models that can accu-  
433 rately capture this observations. Nevertheless, the magnitude of collision and hydrodynamic  
434 stresses in the segregated layers of the bidispersed fluidized-bed are very similar to those in  
435 the corresponding monodispersed fluidized-bed.

436 The results clearly indicate that both macroscopic and microscopic properties of a  
437 monodispersed fluidized bed can be transferred to a segregated bidispersed liquid-solid  
438 fluidized bed. However, in the transition region, while the volume fraction and collision  
439 stresses are always bounded by values in the lower and upper layers, the particle veloc-  
440 ity fluctuations and hydrodynamic stresses are not always monotonically decreasing with  
441 height. This shows that the properties of the transition region cannot be approximated as  
442 simple averages of those properties in the segregated layers.

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#### 465 **Appendix A: Simulation setup of a fluidized bed**

466 For the simulation of FB-Mono-10 [16], three-dimensional simulations are conducted with  
 467  $N_p = 2000$  particles in a rectangular domain. The particles have an Archimedes number  
 468  $Ar = 23600$ . The grid spacing is uniform in the  $x$ ,  $y$  and  $z$  directions and the grid resolution  
 469 is given by  $\Delta x = \Delta y = \Delta z = h = d_p/25.6$ . The rectangular domain has cross sectional  
 470 dimension  $L_x = L_y = 10d_p$  and its length is  $L_z = 60d_p$  with  $256 \times 256 \times 1536$  grid points. The  
 471 time-step size is  $\Delta t = 1.5 \times 10^{-4}$  s, resulting in a maximum advection Courant number of  
 472 0.5 for the six cases simulated. The cases are run with periodicity in the  $x$  and  $y$  directions.  
 473 The pressure is specified at the top boundary as  $p = 0$ , while at the bottom boundary the  
 474 inflow velocity is specified as uniform and given by  $\tilde{U}$ . The primary parameter of interest  
 475 is the particle Reynolds number  $Re_{p,1} = u_0 d_{p,1} / \nu_f$ , where the average upflow velocity at the  
 476 inlet,  $u_0$ , is varied to investigate Reynolds number effects. A total of six simulations were  
 477 conducted with  $0.010 \leq u_0 \leq 0.035$ , giving  $20 \leq Re_{p,1} \leq 70$ .

#### 478 **Appendix B: Evaluation of different approaches in computing wave speed**

479 In this section, we compare three different approaches which are 1) naive, 2) autocorrela-  
 480 tion and 3) dispersion relation in computing wave speed from volume fraction. To separate  
 481 the wave motion from the random noise, we followed the procedure by Ref [16] to recon-  
 482 struct  $\phi(z, t)$  into its low  $\phi_{k < k_{thresh}}(z, t)$  and high  $\phi_{k \geq k_{thresh}}(z, t)$  wavenumber components  
 483 using Fourier transforms with cut-off wavenumber  $k_{thresh}$ . In this paper,  $k_{thresh} = L_b/d_{p,1}$ ,  
 484 where  $L_b$  is the height of the monodispersed layer or each segregated layer in the bidispersed  
 485 fluidized bed. Figure S1(b) shows the reconstructed low wavenumber  $\phi_{k < k_{thresh}}(z, t)$  signal.  
 486 Compared to figure S1(a), the wavelike behavior is more distinct, and an approximate wave

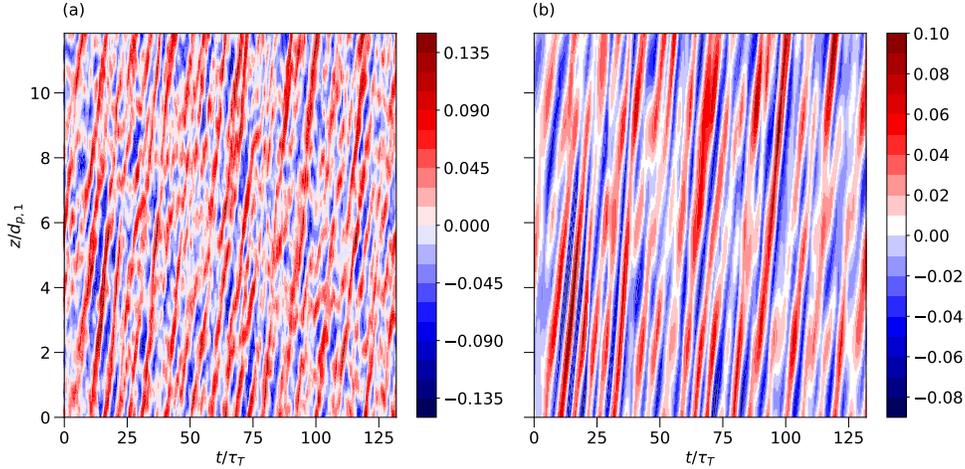


Figure S1. Volume fraction fluctuation  $\phi'$  as a function of time  $t$  and vertical position  $z$  at  $Re_p = 60$  in the lower layer for case FB-Bi-12. (a) unfiltered  $\phi'$ , (b) reconstructed low-pass filtered  $\phi'$ .

487 speed can be computed based on the slope of the features in the  $z - t$  plane.

488 For the naive approach, we approximate the wave speed directly from figure S1(b) by  
 489 computing the average of  $z/t$  for  $t$  that results in the top five largest  $\phi_{k < k_{thresh}}(z, t)$  for each  
 490 respective  $z$ . The naive approach wave speed  $c_{na}$  is formally defined as

$$c_{na} = \frac{1}{N_k N_i} \sum_{k=1}^{N_k} \sum_{i=1}^{N_i} \frac{z_k}{t_{k,i}}, \quad (\text{B1})$$

491 where  $N_i = 5$  is a constant that determines the number of values to be used for each  $z$ ,  $N_k$   
 492 is the number of grid points in the fluidized bed,  $z_k$  is the vertical position and  $t_k$  is the  
 493 value of  $t$  that results in the top  $N_i$  largest  $\phi_{k < k_{thresh}}(z, t)$  at  $z_k$  which is defined as

$$t_k = \arg \max_{\psi \in \phi_{k < k_{thresh}}(z, t), |\psi| = N_i} \sum_{v \in \psi} v, \quad (\text{B2})$$

495 where  $|\psi|$  denotes the number of elements in  $\psi$ .

496 For the two-dimensional autocorrelation approach, we followed the procedure in Ref [43].  
 497 The space-time autocorrelation of  $\phi'$  is defined as  $\langle \phi'(z + \Delta z, t + \Delta t) \rangle$ . By assuming  $\phi'(z, t) =$   
 498  $\phi'(z - ct)$  in the form of a propagating wave and wave speed  $c = \Delta z / \Delta t$ , the autocorrelation  
 499 of  $\phi'$  is reduced to  $(\phi')^2(z, t)$ , appearing as the maximum value in the autocorrelation plot.  
 500 The advantage of this approach is that the dominant wave will be amplified, hence making  
 501 the wave speed approximation more reliable. A detailed validation of this approach can  
 502 be found in Ref [43]. Figure S2(a) shows a typical space-time autocorrelation plot of  $\phi'$ .

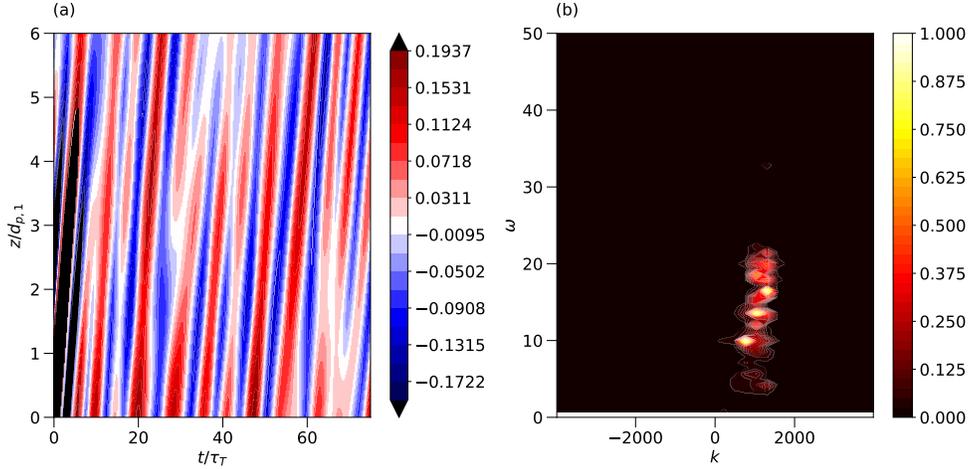


Figure S2. (a) Autocorrelation of the low-pass filtered volume fraction fluctuation  $\phi'$  as a function of time  $t$  and vertical position  $z$  and (b) Energy spectra of the reconstructed volume fraction fluctuation  $\phi'$  as a function of wavenumber  $k$  and frequency  $\omega$  at  $Re_p = 60$  in the lower layer for case FB-Bi-12.

503 Although the wave-like behavior is clearer when compared to Figure S1(b), the wave-like  
 504 bands in our simulations are not as clear as those in Ref [43]. A plausible explanation for this  
 505 lack of obvious wave-like motion is the method of forcing in our simulations. In Ref [43], a  
 506 triply periodic domain is used and vertical forcing is added directly to the flow to balance the  
 507 weight of particles. Our simulations are doubly periodic with inflow and outflow specified at  
 508 the top and bottom boundaries, thus representing a more realistic and perhaps noisier result  
 509 characterized by disturbances propagating through the domain due to boundary effects. To  
 510 approximate the autocorrelation wave speed  $c_{auto}$ , equation B1 is used by computing the  
 511 ratio of  $z$  to  $t$ .

512 For the dispersion relationship approach, we construct the energy spectra of  $\phi'$  using  
 513 the Fourier transform to compare the energy spectra as a function of frequency  $\omega$  and  
 514 wavenumber  $k$ , and then approximate the wave speed with  $c = \omega/k$ . Figure S2(b) shows  
 515 the energy spectra normalized by the maximum value in two-dimensional  $k - \omega$  space. (A  
 516 Peak is defined where the normalized energy spectrum is greater than 0.8 and each peak  
 517 represents a wave speed  $\omega/k$ ). As shown in figure S2(a), three peaks are observed that fall  
 518 on the same line defined by  $\omega = c_{FT}k$ , indicating the dominance of three different wave  
 519 modes propagating at the same speed. The wave speed  $c_{FT}$  is approximated by fitting the

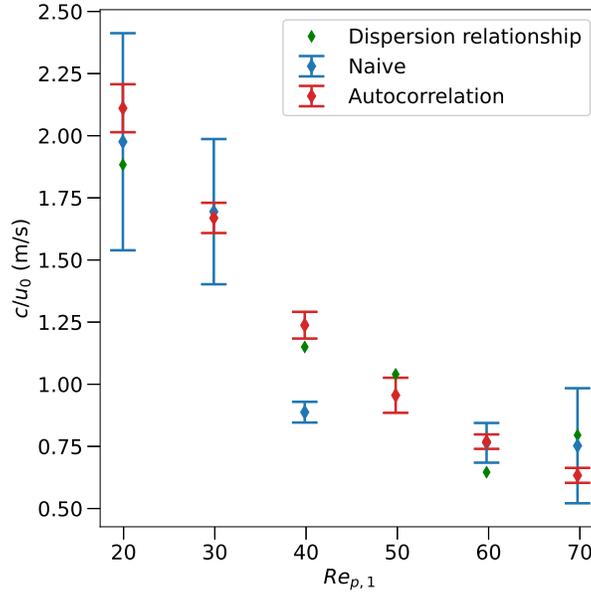


Figure S3. Wave speed derived from different approaches as a function of  $Re_{\rho,1}$  for case FB-Mono-12.

520 line defined by  $\omega = c_{FT}k$  to the three peaks.

521 Figure S3 shows the wave speed computed with different approaches for FB-Mono-12.  
 522 Overall, the wave speed derived from the autocorrelation function has the least uncertainty,  
 523 indicated by the smallest standard deviation. The naive approach gives results with similar  
 524 averages but much larger standard deviations. This is expected because of ineffective noise  
 525 suppression as shown in figure S1(b). Interestingly, the dispersion relationship approach  
 526 gives almost identical results as the autocorrelation approach. However, the main disadvan-  
 527 tage of the dispersion relationship is the need for a large domain. If the domain is small such  
 528 that the wavelength of the wave is greater than the fluidized-bed height, smeared peaks will  
 529 be observed leading to inaccurate results. Since the segregated bed height in cases FB-Bi-12  
 530 and FB-Bi-14 are smaller than the wavelength, the dispersion relationship is less accurate  
 531 for these cases. In the remainder of this work, we adopt the autocorrelation approach to  
 532 compare wave speeds.

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