Pattern formation in pulsed gas-solid fluidized beds – The role of granular solid mechanics

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HIGHLIGHTS

• First successful CFD-DEM approach to model pattern formation in pulsed fluidized beds.
• Simulations reproduce experimental patterns, both qualitatively and quantitatively.
• Solids friction is critical to reproduce regular bubble patterns in deep beds.
• Pattern formation may serve as simple yet robust “fingerprint” to validate simulation approaches.

GRAPHICAL ABSTRACT

ABSTRACT

Under certain conditions, gas-solid fluidized beds are known to develop a structured flow of bubbles when exposed to periodically pulsating air flows. In quasi-two-dimensional beds, periodically rising bubbles form a triangular tessellation in the vertical plane. Bubble nucleation sites at the distributor plate alternate during each cycle. This pattern sets an excellent benchmark for fundamental studies of fluidization. Notably, most common Eulerian descriptions of granular flow do not yet capture this interplay between solid mechanics and fluid-solid momentum exchange, which we show to be instrumental to the dynamic rearrangement of bubbles in a pulsed bed. We report the first successful CFD simulations of structured bubble flows in a deep, quasi-2D geometry using a Eulerian-Lagrangian CFD-DEM framework. Numerical results are in quantitative agreement with experiments. The simulated dynamics reveal that the patterns emerge from the transition of the granular collective behavior between solid-like and fluid-like, which is an outcome of dynamical coupling between gas and particles. The simulated results point out the essential role of solid frictional stresses on inducing and maintaining the formation of bubble patterns. This underscores the value of investigating pulsation-induced patterns as a prime manifestation of the mesoscopic physics underpinning fluidization, and highlights the direction for improving current practices.

1. Introduction

Bubbling gas-solid fluidized bed reactors are widely used in various industrial applications, due to their excellent mixing properties and interfacial heat and mass transfer [1,2]. Their overall performance largely relies on the bubble dynamics: rising bubbles drive the solids circulation and significantly enhance gas-solids contact, improving mixing and transport properties. However, highly nonlinear collective behavior arises from the dissipative collisions between particles, and the seemingly chaotic coalescence and breakup of bubbles. Together, these give rise to

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very complex multiphase flow dynamics [3,4], challenging every aspect of the engineering, design and scale-up of fluidized bed reactors.

Any means to structure the bed hydrodynamics can facilitate engineering fluidized bed reactors. Some intrusive methods, such as distributed gas injectors, internal baffles and electrodes, are used to impose order on a chaotic process by manipulating the distribution of the gas supply and the particle dynamics [5]. However, one of the most effective and non-intrusive ways to structure bubble dynamics is the application of a periodically pulsed gas flow. Pulsating the air supply in gas-solid fluidized beds can impose an ordered pattern to the bubble flow [6]. In a quasi-two-dimensional (2D) geometry, gas bubbles are observed to rise, spaced with a characteristic transversal wavelength \( \lambda \), independent of the system dimensions [7]. They are stacked according to a triangular tessellation, in which bubbles shift positions by \( \lambda/2 \) at each cycle and remain staggered in rows that are vertically aligned to form a structured array (Fig. 1). The resulting configuration is subharmonic, as it constantly recurs at half of the pulsating frequency \( f/2 \). This striking phenomenon not only provides the potential to improve the operation and design of gas-solid fluidized beds, but also excels as a rigorous way to validate computational methods, since the bubble arrangement, as is demonstrated further on, relies on the complex interplay of fluid-solid forces and changes of granular rheology in time and space.

Over the past 20 years, computational fluid dynamics (CFD) have been increasingly used to provide insights into fluidization, while avoiding some of the difficulties of direct experimentation [8–10]. Depending on the required level of detail, two state-of-the-art approaches of modeling gas-solid fluidized systems are prevalent: the discrete element method (denoted as DEM) or Lagrangian–Eulerian approach, and the two-fluid model (denoted as TFM) or Eulerian–Eulerian approach.

Discrete element methods explicitly track and solve every single particle trajectory following Newton’s laws of motion [11,12]. Inter-particle contacts are depicted mathematically in either hard-sphere [13] or soft-sphere fashion [14]. In a dense granular flow, particles encounter many sustained contacts, and multiple collisions occur simultaneously. Since a soft-sphere treatment handles multiple contacts in a more robust way, it is favored for the analysis of granular mechanics [15–17] and local velocity fluctuations [18,19]. Coupling the CFD and DEM methods is computationally expensive. Flow fields are computed on discrete grids with a grid size at least one order of magnitude above the particle diameter [20]. Unfortunately, implementation is still uneconomical for typical industrial units or even pilots, where the characteristic spatial scales of the flow and the particles differ by several orders of magnitude. In this case, the number of particles to be considered in a simulation becomes impractical and the computational load increases dramatically. To this date, a direct CFD-DEM

**Nomenclature**

- \( A \) average of oscillating flow, (-)
- \( B \) amplitude of oscillating flow, (-)
- \( C_d \) particle drag coefficient, (-)
- \( d_{eq} \) equivalent bubble diameter, (m)
- \( d_i \) particle diameter, (m)
- \( d_c \) CFD grid size, (mm)
- \( \epsilon \) restitution coefficient, (-)
- \( E \) Young’s modulus, (Pa)
- \( E_k \) granular kinetic energy, (J)
- \( f \) frequency of oscillating flow, (Hz)
- \( F_c \) inter-particle contact force, (N)
- \( F_t \) inter-phase contact force, (N)
- \( g \) gravitational acceleration, (9.81 m/s²)
- \( G \) shear modulus, (Pa)
- \( I \) moment of inertia, (kg m²)
- \( k_n \) normal spring constant, (N/m)
- \( k_t \) tangential spring constant, (N/m)
- \( m \) particle mass, (kg)
- \( M_k \) interphase momentum exchange, (kg/(m² s²))
- \( n \) number of particle, (-)
- \( N \) natural number, (-)
- \( p \) gas pressure, (Pa)
- \( P \) solid pressure gradient vertical component, (Pa/m)
- \( Re \) particle Reynolds number, (-)
- \( t \) flow time, (s)
- \( T \) torque, (N m)
- \( t_0 \) initial flow time (s)
- \( t_c \) characteristic collision time, (s)
- \( T_s \) solid stress tensor, (Pa)
- \( U_0 \) superficial gas velocity, (m/s)
- \( U_s \) centroid velocity of gas phase, (m/s)
- \( U_{mf} \) minimum fluidization velocity, (m/s)
- \( U_n \) normal particle relative velocity, (m/s)
- \( U_l \) tangential particle relative velocity, (m/s)
- \( U_t \) tangential particle relative velocity, (m/s)
- \( V_c \) cell volume, (m³)
- \( V_i \) particle volume, (m³)
- \( x \) lateral distance from origin (cm)
- \( y \) vertical distance from the distributor (cm)

**Greek symbols**

- \( \beta_d \) drag force coefficient, (kg/(m³ s))
- \( \gamma_n \) normal damping coefficient, (N/m)
- \( \gamma_t \) tangential damping coefficient, (N/m)
- \( \delta_{ip} \) normal particle overlapping, (m)
- \( \delta_t \) tangential particle overlapping, (m)
- \( \varepsilon \) volume fraction of gas phase, (-)
- \( \zeta \) interpolation coefficient, (-)
- \( \varphi \) angle of repose, (-)
- \( \lambda \) pattern wavelength (cm)
- \( \mu_g \) viscosity of gas phase, (Pa s)
- \( \mu_{ip} \) inter-particle frictional coefficient, (-)
- \( \mu_{pw} \) wall-particle frictional coefficient, (-)
- \( \nu \) Poisson’s ratio, (-)
- \( \rho_g \) gas density, (kg/m³)
- \( \rho_s \) solids density, (kg/m³)
- \( \sigma_{ip} \) deviator stress tensor of gas phase, (Pa)
- \( \phi \) initial volume fraction of solid phase, (-)
- \( \phi_0 \) rotational velocity of particle, (rad/s)
- \( m^* \) effective mass between two contacting particles:
  \( \frac{1}{m^*} = \frac{1}{m_1} + \frac{1}{m_2} \)
- \( R^* \) effective radius between two contacting particles:
  \( \frac{1}{R^*} = \frac{1}{R_1} + \frac{1}{R_2} \)
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implementation for granular flows is limited to the scope of fundamental studies [12,21,22]. However, discrete element methods have become increasing popular in recent years, even for applied studies. Other Lagrangian approaches, such as hybrid codes [23,24] and coarse-grained methods [25] serve to tackle large-scale simulations by representing the dispersion of the particulate phase with a lower number of discrete entities or parcels, each accounting for a ratio of the mass flow equivalent to many real particles.

Eulerian descriptions are suited to tackle large-scale multiphase systems [4,26]. In granular flows, a two-fluid model (TFM) describes gas and solid phases as interpenetrating continua, which substantially reduces the computational demand and allows one to model the macroscale dynamics of particulate phases more inexpensively. Nevertheless, omitting the description of local interactions gives rise to new transport equations and the need for constitutive closures, defining a solids viscosity and pressure. In analogy to the study of molecular gases, the well-established kinetic theory of granular flow (KTGF) is often implemented within the TFM framework to describe non-linear solid stresses under the assumption that particle interactions occur through binary, instantaneous collisions in the so-called viscous regime [27]. Naturally, KTGF is not applicable when particles are closely packed, causing multiple, simultaneous contacts. For that reason, a Eulerian description based on the KTGF greatly underestimates the effective solid stresses in dense granular flows. In order to mitigate these errors and bridge the solid-like and liquid-like behavior of granular matter, the frictional solid stresses are computed by coupling the KTGF with soil mechanics based frictional stress models that compensate for the deviations at low void fractions, the so-called plastic regime. As a result, the simulation of solids circulation and bubbly flows in dense systems becomes greatly reliant on frictional stress correlations [28–30].

Modeling a periodically pulsed bed constitutes a particular challenge, as the rheology changes in time, and dense regions develop locally. Only a few computational works have been reported in this direction. The first reported modeling attempt used a simple one-dimensional Particle Array Model (PAM) [31], in which particles form a vertical string [32]. The authors observed highly ordered particle movement by applying oscillatory gas flows, and the most regular movement was captured at a pulsation frequency of 4–5 Hz. Nonetheless, omitting the description of local interactions of the TFM available in commercial packages are unable to reproduce the structured bubble flows in a quasi-2D pulsed bed. It follows that standard practices must, to some extent, incorrectly describe solid circulation and residence time. It especially demonstrates that the details of the model matter in reproducing the experimental bubble patterns.

In this work, we progress to simulate the pulsed fluidization of granular matter, and evaluate the performance of CFD-DEM approaches in reproducing a dynamic bubble pattern. We demonstrate for the first time that in a deep, quasi-2D geometry, the emergence of structured patterns can be reproduced quantitatively using several conventional CFD-DEM implementations. This provides a powerful tool to study the role of granular rheology in the stabilization of coherent bubble flows and may serve as a benchmark for the future development of advanced continuum models for fluidized granular flow.

2. Model settings

Simulations of a quasi-2D bed are conducted with the open-source code CFDEM, version 3.1.0, which employs a four-way coupled solver [36]. The following sections summarize the governing transport equations and sub-models used in this work.

2.1. Governing equations

The gas phase dynamics are governed by the conservation of mass (Eq. (1)) and linear momentum (Eq. (2)), assuming the gas phase to be incompressible and isothermal, at ambient conditions, thus exhibiting constant viscosity \( \mu_g \):

\[
\frac{\partial (\rho_g U_g)}{\partial t} + \nabla \cdot (\rho_g U_g U_g) = 0
\]  

(1)

\[
\frac{\partial (\rho_g U_g)}{\partial t} + \nabla \cdot (\rho_g U_g U_g) = -\rho_g \frac{\partial P}{\partial x} + \nabla \cdot \tau_g + \rho_g g + M_g
\]  

(2)

The gas stress tensor is modeled with the Newtonian strain-stress relation:

\[
\tau_g = \mu_g (\nabla U_g + \nabla U_g^T)
\]  

(3)

The motion of all particles individually is resolved using DEM. For each single particle of mass \( m \), its translational velocity \( U_p \) and rotational velocity \( \omega_p \) follow Newton’s laws of motion:

\[
m \frac{dU_p}{dt} = F_c + F_l + mg
\]  

(4)

\[
l \frac{d\omega_p}{dt} = T
\]  

(5)

where \( F_c \) and \( F_l \) are the forces representing inter-particle collisions and gas-solid interaction, respectively; \( g \) is the gravitational acceleration; \( T \) is the torque induced by the tangential contribution of

\[ F_c, F_l, m, g, T \]
the contact, and I is the moment of inertia. The normal and tangential inter-particle contacts are modeled using Hertzian spring-dashpot contact theory [11], expressed as a linear combination of a spring contribution and a damping contribution:

\[ F_n = k_n \delta_n - 2 \sqrt{\frac{5}{6}} \sqrt{\frac{2}{2}} k_{nm} \mathbf{U}_n \]  

\[ F_t = k_t \delta_t - 2 \sqrt{\frac{5}{6}} \sqrt{\frac{2}{2}} k_{tm} \mathbf{U}_t \]  

\[ k_n = \frac{4}{3} E' \sqrt{R' \delta_n} \]  

\[ k_t = 8 G' \sqrt{R' \delta_t} \]  

Here, \( E' \) and \( G' \) stand for the effective Young’s modulus and shear modulus, respectively; \( R' \) is the effective radius; \( \delta_n \) and \( \delta_t \) are the normal and tangential geometric overlap between the paired particles; \( k_n \) and \( k_t \) are the normal and tangential spring stiffness.

In addition, the tangential force is finite, and it is common practice to impose Coulomb’s criterion when the tangential force exceeds the maximum static friction [37,38]; therefore, the tangential force is limited to be smaller or equal than the maximum static friction:

\[ F_t \leq \mu_s F_n \]  

where \( \mu_s \) is the friction coefficient. Furthermore, in Eqs. (6) and (7):

\[ \beta = \frac{\ln(e)}{\sqrt{\ln^2(e) + \pi^2}} \]  

where \( \beta \) is defined as a function of the coefficient of restitution, \( e \). The particle-wall collisions are modeled assuming an extremely large value for the mass of the wall when computing the effective material properties.

2.2. Interphase force

In the context of these simulations, the interphase momentum exchange is considered to be dominated by the drag and buoyancy forces, whereas other contributors, such as lift forces and virtual mass forces can be considered negligible [39]. Drag is modeled as a function of the relative velocity of the gas to the solids. It is obtained empirically and calculated individually for each particle. The fluid-particle interaction \( F_t \) exerted on the particle \( i \) is expressed by:

\[ F_i' = -V_i \nabla p + \frac{V_i \rho_d}{1 - \varepsilon} (\mathbf{U}_g - \mathbf{U}_i) \]  

where \( V_i \) is the volume of the particle, \( p \) is the gas pressure, \( \rho_d \) is the drag force coefficient per unit volume of suspension, \( \varepsilon \) is the voidage, and \( \mathbf{U}_g \) and \( \mathbf{U}_i \) are, respectively, the centroid gas and individual particle velocities. A shape correction factor would have to be introduced when using non-spherical particles.

Four-way coupling of the gas-solid momentum exchange follows the same assumptions. For a monodisperse solid phase, the gas-solid momentum transfer term \( M_g \) reads:

\[ M_g = \frac{1}{V_c} \sum_{i=1}^{N} \zeta_i V_i \rho_d \frac{1}{1 - \varepsilon} (\mathbf{U}_g - \mathbf{U}_i) \]  

where \( V_c \) is the cell volume and \( N \) is the number of particles in the present cell. \( \zeta_i \) is an interpolation factor correcting the contribution of each particle based on its position relative to the center of the cell. The drag coefficient \( \rho_d \) is modeled according to Gidaspow’s empirical correlation [40], which combines the correlations of Wen and Yu [41] and Ergun [42] for dilute and dense flows.

In the very dilute regions of the bed, where \( \varepsilon > 0.8 \):

\[ \beta_d = \frac{3}{4} C_d \frac{\rho_d \parallel (\mathbf{U}_g - \mathbf{U}_i)(1 - \varepsilon)}{d_i} \]  

\[ C_d = \frac{24}{R \alpha} \left[ 1 + 0.15 (\varepsilon \alpha)^{0.687} \right] \]  

in which the relative Reynolds number is defined as:

\[ \alpha = \frac{\rho_g}{\mu_g} \parallel (\mathbf{U}_g - \mathbf{U}_i) \| d_i \]  

On the other hand, when \( \varepsilon < 0.8 \), the drag coefficient takes the following form:

\[ \beta_d = \frac{150}{\varepsilon d_i^2} \left( 1 - \varepsilon \right)^2 \frac{\rho_d}{\mu_g} \parallel (\mathbf{U}_g - \mathbf{U}_i) \parallel d_i \]  

2.3. Experimental setup

The setup is schematically shown in Fig. 2. Experiments were conducted in a rectangular (45 cm wide × 1 cm thick) quasi-2D cell made of Plexiglas equipped with a 3 mm thick sintered metal distributor plate Grade 07 (BK 10.30.07, Sintertech). Spherical soda-lime glass beads, with an average diameter of 238 μm, were fluidized using dry air at ambient conditions. The initial bed height was 4.5 cm. The air was pulsed periodically with a superficial

![Fig. 2: Schematic of the experimental setup. The solenoid valve is linked to a computer to control the flow rate. PVC pipes with a diameter of 1.5 cm were used to connect the system.](image-url)
velocity \( U_0 \) given below in terms of the dimensionless mean flow \( A \) and amplitude \( B \):

\[
U_0/U_{mf} = A + B \sin(2\pi ft)
\]  

The minimum fluidization velocity, \( U_{mf} \), was measured experimentally to be 0.041 m/s.

The wavelength of the patterns is determined by the characteristics of the oscillating flow and is independent of the bed dimensions [7]. Therefore, in order to minimize the computational load, the simulations were conducted in a \( 10 \times 0.2 \times 10 \) cm quasi-2D rectangular domain. To validate this, an additional simulation conducted in a \( 15 \times 0.2 \times 10 \) cm domain was carried out as well, and, indeed, reproduced a similar pattern wavelength. This domain size is sufficient to encompass the generation of a row of two bubbles, and it can be used to demonstrate the nucleation process and stabilization of a pattern.

2.4. Computational setup and numerical implementation

Table 1 summarizes the computational parameters. Zero-flux boundary conditions were assigned to the front and rear walls to eliminate any effects on the gas phase. Poisson’s ratio, \( \nu \), was set to 0.22, and the coefficient of restitution, \( e \), of the glass beads in inter-particle and wall-particle collisions was set to 0.97 [43]. According to the verified practices of slowly bubbling fluidized beds [43,44], an artificially small Young’s modulus (10 MPa) was used for the glass beads to avoid a prohibitively small DEM time step. When non-cohesive Geldart B particles with a spring stiffness above 800 N/s are used [34,44,45], the precise value of the spring stiffness has no significant influence on the bubbling behavior [46]. By piling the glass beads on a horizontal surface, the angle of repose, \( \theta \), was measured experimentally and the friction coefficient between particles, \( \mu_{sp} \), was set to 0.35 according to the approximation \( \tan(\theta) \approx \mu_{sp} \) obtained from the Mohr–Coulomb criterion; it was reduced to 0.1 for particle-wall collisions, \( \mu_{ew} \).

Table 1: Settings of the CFD-DEM simulations.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Solid phase</td>
<td></td>
</tr>
<tr>
<td>Particle density, ( \rho_p )</td>
<td>2500 kg/m(^3)</td>
</tr>
<tr>
<td>Mean particle diameter, ( d_1 )</td>
<td>238 ( \mu )m</td>
</tr>
<tr>
<td>Young’s modulus, ( E )</td>
<td>10 MPa</td>
</tr>
<tr>
<td>Restitution coefficient, ( e )</td>
<td>0.97</td>
</tr>
<tr>
<td>Poisson’s ratio, ( \nu )</td>
<td>0.22</td>
</tr>
<tr>
<td>Inter-particle friction</td>
<td>0.35</td>
</tr>
<tr>
<td>coefficient, ( \mu_{sp} )</td>
<td></td>
</tr>
<tr>
<td>Particle-wall friction</td>
<td>0.1</td>
</tr>
<tr>
<td>coefficient, ( \mu_{ew} )</td>
<td></td>
</tr>
<tr>
<td>Gas phase</td>
<td></td>
</tr>
<tr>
<td>Gas density, ( \rho_g )</td>
<td>1.225 kg/m(^3)</td>
</tr>
<tr>
<td>Gas viscosity, ( \nu_g )</td>
<td>( 1.8 \times 10^{-5} ) Pa s</td>
</tr>
<tr>
<td>Common</td>
<td></td>
</tr>
<tr>
<td>Bed width</td>
<td>10 cm</td>
</tr>
<tr>
<td>Initial bed height</td>
<td>4.5 cm</td>
</tr>
<tr>
<td>Simulation domain</td>
<td>( 50d_1 \times 1d_1 \times 50d_1 )</td>
</tr>
<tr>
<td>( W \times T \times H )</td>
<td></td>
</tr>
<tr>
<td>CFD grid size, ( d_1 )</td>
<td>2 mm</td>
</tr>
<tr>
<td>Side wall boundary condition</td>
<td>No-slip for the gas phase</td>
</tr>
<tr>
<td>Inlet boundary condition</td>
<td>Superficial velocity: ( U_0/U_{mf} = 2.64 + 2.14 \sin(2\pi ft) )</td>
</tr>
<tr>
<td>Outlet boundary condition</td>
<td>Constant pressure (101325 Pa)</td>
</tr>
<tr>
<td>Time step</td>
<td>Solid phase: ( 1 \times 10^{-6} ) s, Gas phase: ( 1 \times 10^{-8} ) s</td>
</tr>
</tbody>
</table>

2.5. Analysis of the bubble properties

During the patterned state, the properties of bubbles are computed when the distance from the center of the bubble to the distributor is between 2.5 and 3.5 cm. In simulations, the cells of bubbles are recognized by imposing a sharp threshold (\( \varepsilon > 0.8 \)). Considering a quasi-2D geometry, the area of a single bubble is assumed to be equal to the sum of the areas of connected cells containing void space. All the bubbles are assumed to have a rounded shape to estimate their equivalent bubble size. The pattern wavelength is measured as the distance between each pair of bubbles, which nucleate during the same pulse. Nevertheless, the above analysis loses its meaning when the flow of bubbles becomes chaotic.

3. Results and discussion

3.1. Experimental and simulated pattern formation

Experiments were conducted under sets of conditions corresponding to the computational domain to ensure the opportunity for direct validation. In both cases, a sinusoidal flow oscillating between 0.5\( U_{mf} \) and 4.78\( U_{mf} \) and with a frequency of 5 Hz or 7 Hz was introduced. In both frequency sets, the instability emerges spontaneously in the experiments after only a few periods. The bubbles are nucleated in a structured array (Fig. 4a), which is then sustained, flowing upwards during an entire cycle (see Supplementary Material for videos of experimental patterns). The bubble nucleation sites in the subsequent cycle emerge in between the bubble locations of the previous array, thus leading to stacked rows in which the bubbles alternate positions. In deeper beds, this type of arrangement develops into a recognizable triangular tessellation pattern in the vertical plane, as earlier work from our group has shown [5,7]. As expected, both the wavelength of the pattern, corresponding to the horizontal separation of the bubble nucleation sites, and the bubble size decrease with increased pulsation frequency, Table 2.

After a few periods, CFD-DEM simulations under the same pulsed flows conditions, Table 1, lead to a dynamic, subharmonic bubble pattern, in excellent agreement with the experiments (Fig. 4b). The bubble nucleation sites near the distributor rearrange dynamically after each cycle in the same way as in the experimental case, forming two stacked rows where bubble positions are shifted every cycle by half a wavelength in the horizontal direction. The computational pattern is stable and recurs at half of the
pulsation frequency. To our knowledge, the data shown provide
the first unambiguous reproduction of dynamic bubble patterns
in quantitative agreement with experiments in terms of the
arrangement, average bubble size and wavelength, Table 2 (see
Supplementary Information for videos of simulated patterns).

3.2. Description of the bed dynamics during a patterned state

CFD-DEM simulations can provide greater insight into the
mechanism of pattern formation as well. In this section, we look
into the gas and solid phase dynamics of this newly defined
structured bubble flow, and the key contributors to its stabiliza-
tion. To avoid redundancy, the following analysis features only a
few periods of the recurrent bubble structure and a different time
during a pulse period ($T$) once it has reached a stable state
after $\sim 3$ s. As a periodic phenomenon, it is convenient to use the
phase angle, $\phi$, to describe the pattern evolution during any single
period of the oscillating gas flow. The phase angle is defined as
$\phi = 2\pi(t - t_0)/T$ with an initial flow time $t_0 = NT$, where $N$ is a
natural number.

3.3. Dynamics of the gas phase

Fig. 5 shows the recurrent field of the gas pressure, coupled
with the rising bubbles. The pressure drop over the bed fluctuates
according to the oscillatory gas flow, peaking at $\phi = \pi/2$.

Table 2
Comparison between experimental and computational results. The conditions are listed in Table 1.

<table>
<thead>
<tr>
<th>Pulsed frequency (Hz)</th>
<th>Bubble size, $d_{eq}$ (cm)</th>
<th>Wavelength $\lambda$ (cm)</th>
<th>Minimum fluidization velocity, $U_{mf}$ (m/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>5 Hz</td>
<td>Experiment 2.5 ± 0.2</td>
<td>6.5 ± 0.6</td>
<td>0.041</td>
</tr>
<tr>
<td></td>
<td>CFD-DEM 2.2 ± 0.3</td>
<td>6.4 ± 0.2</td>
<td>0.044</td>
</tr>
<tr>
<td>7 Hz</td>
<td>Experiment 1.6 ± 0.2</td>
<td>5.6 ± 0.4</td>
<td>0.041</td>
</tr>
<tr>
<td></td>
<td>CFD-DEM 1.3 ± 0.6</td>
<td>5.1 ± 0.8</td>
<td>0.044</td>
</tr>
</tbody>
</table>

Fig. 3. (a) Sensitivity analysis on CFD grid size for CFD-DEM simulations. The static solid packing becomes size independent when the grid size is above 2 mm; (b) Snapshot of the 2 mm cubic CFD grid used for simulations. The thin black lines represent the mesh grid.

Fig. 4. Snapshots for (a) experimental, (b) CFD-DEM patterns in a quasi-2D bed with an oscillating airflow with velocity $U_0/U_{mf} = 2.64 + 2.14\sin(2\pi t)$. The snapshots were taken from consecutive cycles. The bubble nucleation sites alternate every cycle, shifting half of the wavelength $\lambda$ in the transversal direction.
When traveling more slowly than the airflow in the emulsion phase, gas bubbles represent preferable paths for the air to channel through granular media [3]. In the CFD-DEM simulations, the air streamlines converge into the rising bubbles (Fig. 6), which originates from an uneven pressure distribution over the lower domain (Fig. 5). Two regions are clearly differentiated. Due to the lower pressure drop within the bubbles, the area in the wake behind them exhibits a higher air velocity and pressure drop, while the area of high pressure extends further up in between the bubbles.

3.4. Dynamics of the particulate phase

During every oscillation cycle, the superficial velocity of the air falls below $U_{mf}$, causing the particulate phase to be temporarily de-fluidized in the valley of the sinusoidal signal. At this moment, the existing bubbles continue to rise slowly, maintaining their shape from collapsing. The bubble motion drives the displacement of the solids on either side; solids travel downward, then converge and come into contact vigorously within the wake of each bubble (Region 1 in Fig. 7) to eventually reach a quasi-static state. In contrast, the solids in the region between the bubbles (Region 2 in Fig. 7) tend to rise, but exhibit much less movement and undergo a smaller number of collisions.

At $\varphi = 0$, the bulk of the granular matter, including both Region 1 and 2, is still de-fluidized, densely packed with a velocity for the solids approaching zero (Fig. 8a). As the air velocity increases in a new cycle, the system becomes partially fluidized, up to the point of bubble nucleation ($\varphi \sim \pi/4$). During the process, the solids within Region 1 remain static and densely packed, due to a large number of collective contacts and the effect of frictional stress, which suppresses the rise of the particles. The solids within Region 2 respond more rapidly to the change in air velocity and expand to create a new bubble nucleation site (Fig. 8b).

In a pair of rising bubbles, sustained local multi-particle contacts are generated by the circulation of solids into the bubble wake, leading to long-range, large solid stresses at the interface between the bubble and the emulsion. Fig. 9 displays the computed solid pressure, showing significant compressive yields in the wake of a rising bubble. Such a high load explains the restricted mobility of the particles beneath the bubbles; the resulting adverse force to the lifting drag explains a slower response to expansion in the next cycle of increasing air velocity, compared to the central Region 2 in Fig. 7. The solid pressure gradients in both regions

Fig. 5. Snapshots for simulated bubble pattern profiles within a single period of gas pulsation. Gas pressure fields alternate in conjunction with the bubbles. The snapshots were taken from two consecutive periods of the periodically pulsed inlet flow, between 4.2 and 4.4 s flowtime. The blue lines represent isobars for the absolute gas pressure $P$ at 101500 Pa, 101600 Pa, 101800 Pa and 101900 Pa, respectively, from the top to the bottom. The insets schematically show the velocity of the pulsed gas flow at the corresponding phase angle, during one period.
differ approximately by a factor of 3 when the maximum load conditions are reached (Fig. 10). Identifying not only the rheological behavior of the plastic regime, but also the point of transition is of critical importance for TFM (Eulerian-Eulerian) simulation frameworks [15]. In this case, DEM can explicitly solve the stress transmission and demonstrate that the formation of a void is
periods of the periodically pulsed inlet flow. 

Fig. 10. Time series of the vertical component of the average solid pressure gradient, $\nabla P_s$, in the two out-of-phase regions (shown in Fig. 7) for CFD-DEM. The data are sampled from five consecutive pulse periods. The solid pressure is reconstructed using the virial theorem [15] for the CFD-DEM simulations.

The stresses imposed on the bottom region of the bed near the distributor are gradually reduced as the granules become fluidized (Fig. 10). In response to the spatial distribution of the solid pressure gradients, a longitudinal wave-shaped void initially spans the entire distributor and then splits into two alternating bubble nucleation sites when the air velocity starts to decrease (see Supplementary Material for videos of the simulated pattern). The newly formed nucleation sites appear along two vertical axes equidistant to the horizontal position of the previous set of rising bubbles. The process recurs, and hereby gives rise to a complete subharmonic pattern. In wider and deeper beds, the same sequence expands into any number of bubbles stacked in line at a given wavelength and several stacked rows in a triangular tessellation.

4. Discussion

Common CFD-DEM implementations were shown to successfully reproduce, in a quantitative manner, the dynamic bubble patterns observed in a small, quasi-2D fluidized bed. Based on the above observations, the emergence of patterns is associated with the oscillation of granular matter over the frictional packing limit during part of each cycle, thus alternating the granular collective behavior between fluid-like and solid-like. This transition is known to create a dramatic change in non-linear solid stress. Particles become fluidized during the formation of the bubbles, showing a sharp decrease in the solid stress within the bubble wake (Fig. 10). However, due to the circulation of the solids in the wake of the bubbles and the following half-cycle of decreasing air velocity, solids concentrate and remain in a dense, quasi-static form when approaching jamming or transition to the plastic regime. This phenomenon gives rise to a set of non-linearly growing stresses that, collectively, arrange bubbles into a regular pattern. A change in frequency of the perturbation affects the size of the bubbles and, correspondingly, adjusts the characteristic wavelength, since it alters the temporal and spatial scales of the local solids circulation.

The data presented here show that the granular solid mechanics play an essential role in the emergence of structured bubble patterns and their stability. The effects of friction are non-local: they span over long distances, which is fundamental to reach a balance in the upwards propagation of bubble patterns. However, when friction is eliminated and the solids become mobile in the wake of the bubbles, a deep pulsed bed fails to reach a structured state, as shown in Fig. 11. Instead, solids circulate widely, driven by the motion of the bubbles over time. By omitting frictional stresses in the dense region, the model fails to identify the correct jamming point; as a result, fluid dynamics largely dominate over the role of solid mechanics.

In a continuum approach, the description of the granular rheology requires a complex numerical treatment and constitutive equations to reconcile the stress-strain relations over a full range of void fractions, spanning from viscous to transitional and plastic regimes. Advanced models with a dynamic threshold for the jamming point are not yet widely explored in the fluidization community. Using a discrete treatment for the particulate phase allows us to compute stresses and strains by resolving force balances in all individual contacts and, thus, explicitly solve the evolution of rheological properties across the bed. The data shown here indicate that the stabilization of the patterns, that is, the alternation of nucleation sites, is related to the compressive stress generated in the wake of bubbles in the plastic regime. Our previous work has shown that the commonly used continuum models, including the most used correlations of frictional stresses, fail to capture this phenomenon [35]. Therefore, an accurate description of the jamming transition and of the rheology of the transition and dense regimes must be critical to depict when and where bubbles form under different conditions. Classic KTGF assumes collisions to be instantaneous, binary and frictionless below the frictional packing limit, and, thus, it cannot be expected to hold around the bubble wakes characterized by enduring contacts and long-range force chains [16]. Similar challenges occur when modeling sandy piles, an hourglass and U-tubes, where static inter-particle friction is the dominant element [52]. It is also well known that the use of frictional stress models is essential to describe the bubbling behavior in fluidized beds [29,53], particularly when using a low superficial velocity. However, the commonly employed expressions are still highly empirical and provide predictions of frictional stresses that can vary by orders of magnitude [28,54]. Nevertheless, many classical implementations have been validated simply by the correct description of bubble properties, without ensuring the proper

Fig. 11. Snapshots of simulated profiles of the flow pattern within a period of gas pulsation when eliminating particle friction. The snapshots are taken from consecutive periods of the periodically pulsed inlet flow.
description of the solids circulation around the bubbles, which impact the entire bed hydrodynamics and, thus, may affect processes like reactions, drying and coating. Over the last few years, increasing efforts have been devoted to improving the account of friction in modeling dense granular flows. As mentioned before, advanced frictional stress models, based on rheological principles, are currently being developed to bridge the transition between the viscous and the plastic regime in dense granular flows. Fluidized beds are further complicated by fluid-solid interactions, leading to very complex dynamics. One cannot even assume that granular matter necessarily behaves as an isotropic system, while these fundamental approaches are still in an early stage. They will, undoubtedly, pave the way towards more accurate kinetic descriptions of fluidization. This work contributes to this exciting new area by linking the effects of friction in the mesoscopic granular dynamics to a macroscopic phenomenon in fluidization, using microscale models to account for the granular rheology.

5. Conclusions

In this work, we have demonstrated that a Eulerian-Lagrangian approach (CFD-DEM) is able to successfully represent the physics underlying pattern formation in quasi-2D, pulsed, bubbling, gas-solid fluidized beds. Computational studies reveal the importance of the coupling between gas and solid phase when reaching a patterned state. The large and sustained solid stress, arising from a dynamic transition of the solid phase between solid-like and fluid-like behavior, leads to a transverse shift in bubble nucleation site at each cycle. Therefore, accounting for the solid mechanics is necessary to capture the sustained, structured bubble patterns witnessed experimentally. The deficiencies of the continuous, Eulerian-Eulerian approach in describing dense frictional flow confront us with particular challenges when simulating pattern formation in a quasi-2D geometry.

The different results from these two modeling approaches also highlight the ability of pulsation-driven pattern formation to be used as an excellent benchmark for validating implementations of multiphase flow models that are used in computer simulations of fluidization. A robust model for the solid phase is required, not only to represent the granular flow properly in the viscous and plastic regimes, respectively, but also, more importantly, to bridge both kinetic and frictional contributions over the transition regime. Combined with the CFD-DEM approach, these results can serve as a reliable, quantitative reference for future developments of frictional stress models for applications on a macroscopic scale.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.cej.2017.05.152.

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