Anomalous Diffusion in a Bench-Scale Pulsed Fluidized Bed

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Abstract

We present our analysis on micro-rheology of a bench-scale pulsing fluidized bed, which represents a weakly confined system. Non-linear gas-particle and particle-particle interactions resulting from the pulsed flow are associated with harmonic and sub-harmonic modes. At the meso-scale, periodic bubbling pattern is produced, while particle-scale measurements reveal anomalous and non-ergodic diffusion possibly associated with a spatial and temporal dependence. The ensemble-averaged mean-squared displacement is characterized by a non-unique exponent resulting from a combination of stochastic processes. There is an evident disparity with its time-averaged analogue indicating weak ergodicity breaking, which along with ageing represents the non-stationary nature of underlying dynamics. The spread of individual time-averaged displacements is non-Gaussian, asymmetric and has a finite trivial contribution accounting for immobile particles.
I. INTRODUCTION

Multiphase flows exhibit a range of spatio-temporal scales arising from complex non-linear dynamics [1–6]. Fluidization is a notable example, in which particles are suspended by an incoming stream of fluid, whereby they exhibit fluid-like behavior. Travelling kinematic waves in the form of bubbles representing spatial inhomogeneities in solids concentration are common in such systems. In particular, pulsed fluidized beds (PFBs) are characterized by a repeatable bubbling pattern resulting from dynamical structuring and suppression of chaos compared to fluidized beds having a non-perturbed inflow. PFBs have shown improved hydrodynamics by reducing or eliminating channeling or clumping of particles, and enhanced heat and mass transfer properties while being non-intrusive. Studies in the past have primarily reported on the meso-scale and macro-scale observations in PFBs [7–12]. Persistent interactions between dynamical modes are present at different scales, and particle-level description is pivotal in elucidating some of the observed features. Particles evolve depending on a number of factors including external forcing, momentum exchange with the carrier-phase and neighbors, material properties and confines of the system. It is intuitive that in the presence of localized features, different particles have different driving mechanisms. For instance, particles in the vicinity of bubble wakes experience a greater acceleration compared to those near the distributor or other quasi-static zones. We use the term quasi-two-dimensional (quasi-2D) to describe PFB used in this study, since the extent in the depth-wise direction is comparable to the size of bubbles, further verified by high-speed videos which reveal their span.

Previous studies [13–17] have examined velocity fluctuations and reported deviations from ideal Brownian motion. The simplistic assumption of Maxwellian distribution breaks down quite easily in multi-particle systems, and results in anomalous diffusion described by,

$$\langle x^2(\Delta) \rangle \sim \Delta^\gamma$$  \hspace{1cm} (1)

The process is sub-diffusive for $\gamma < 1$ and super-diffusive for $\gamma > 1$, both of which are observed in nature and engineering applications [18–25], while $\gamma = 1$ describes normal Brownian diffusion. There also exist possible flow scenarios which cannot be described by a unique value of $\gamma$ and involve transition of regimes discussed above. Different sources of anomalous diffusion have been studied in the past which include Continuous-Time Random Walk (CTRW), Frac-
tional Brownian Motion (FBM) and the motion governed by Fractional Langevin Equation (FLE), Scaled Brownian Motion (SBM), Transport on a Fractal Support, and Heterogeneous Diffusion Process (HDP). Previous analyses also include combining these parent processes such as CTRW-FLE [26] and SBM-HDP [27], the latter was termed Generalized Diffusion Process (GDP), where diffusivity varies in space and time as follows,

\[ D(x, t) \sim (1 + \beta)D_0|x|^{\alpha}t^{\beta} \]  \hspace{1cm} (2)

The above equation combines spatial and temporal dependence from the underlying HDP and SBM respectively. GDP is sub-diffusive \((\gamma < 1)\) when \(\alpha > 2\beta + 4\), and super-diffusive \((\gamma > 1)\) otherwise. Based on the physics of PFB as explained in our previous study [12], we hypothesize the system hosts a combination of parent processes discussed above, as will be shown in the remainder of this article. We also study the effect of ageing i.e., the time lapsed after initializing an experiment. It must be noted that PFB does not represent confinement in a strict sense. Boundaries or walls are present which reflect particles after inelastic collisions in the lateral direction, while the streamwise transport is governed by balance between drag exerted by the carrier-phase and gravity. Hence, we use the term weak confinement to describe PFB.

II. EXPERIMENTS

The experiments were performed using a quasi-2D geometry shown in Figure 1. The bed was filled with 18g glass particles having a Sauter mean diameter of 394\(\mu m\) and a density of 2.5\(g/cc\), classified under Geldart Group B [28]. Flow rate at the inlet is pulsed in the form of a sine wave,

\[ Q(t) = A + B \sin(2\pi ft) \]  \hspace{1cm} (3)

where, the base flow rate \(A = 2.6l/min\) is set such that the velocity is higher than the minimum fluidization velocity, \(U_{mf}\) which denotes the minimum velocity required to support the weight of solids. Details regarding the measurement of \(U_{mf}\) can be found in Vaidheeswaran et al. [29]. The amplitude B is set to 2.1\(l/min\). In this study, we analyze the dynamics of particulate phase under two pulsing frequencies, \(f=4hz\) and \(6hz\). A fractal distributor was 3-D printed (Figure 1) using a high-precision ultraviolet curing printer. High-speed videos
were captured at 300hz over a duration of 20s using a 120mm Nikon lens and Fastex IL5L sensor, and the unit was back-lit using an LED light source. The resulting spatial resolution was 0.71mm X 0.71mm. Glass particles were tracked using an in-house code, PTVResearch [30] and optical distortions were removed using a calibrated grid and polynomial de-warping technique [31]. Individual tracks were determined by Optical Flow Equations, and the outliers were detected based on proper orthogonal decomposition technique [32]. Further details regarding the experiments have been avoided for the sake of brevity, and can be found in Higham et al. [12].

FIG. 1: Schematic of the PFB set-up used in this study. The zoomed-in image and its inset show the frontal view of fractal distributor and its rough sketch (not drawn to scale).

Images reveal meso-scale response of the system to pulsing conditions (Figure 2). At f=4hz, bubbles alternate from wall to wall, while the pattern changes to a bubble at the center and two simultaneous bubbles along the wall at 6hz. There is an active interaction between harmonic and sub-harmonic modes associated with each configuration as explained in our previous study [12]. Force chains are repeatedly formed and broken depending on f, and the rate at which this occurs determines the bubbling pattern having a characteristic wavelength $\lambda$. $\lambda$ representative of bubble size appears to drop while changing f from 4hz to 6hz, and the bubbles are shifted by $\lambda/2$ between successive cycles.
FIG. 2: Bubbling pattern in the PFB at $f=4 \text{Hz}$ (top) and $6 \text{Hz}$ (bottom).

III. RESULTS

Arbitrarily chosen particle tracks are shown in Figure 3 indicating a non-uniform diffusion coefficient in the PFB. Trajectories evolve depending on the initial location as well as appear to have a temporal dependence. We also notice a few tracks which are transported over much shorter distances compared to the rest. Even if a single particle track is considered, depending on the time instance, the motion is altered significantly. If we were to describe using meso-scale features, particles in the wake of bubbles are associated with longer steps, while they undergo much shorter displacements away from the vicinity of such kinematic waves. Also, the motion is confined by walls in the lateral direction, while the balance between gravity and inter-phase drag determines the streamwise transport. Particle trajectories are influenced by carrier-phase depending on their location, a mechanism neither trivial nor explicitly modeled in most of the studies describing anomalous diffusion. In the framework of kinetic theory, the effect due to drag was recently included in the generalized Langevin equation, \cite{33}, and further efforts are required to substantiate this approach.

Next, we look at the autocorrelation, $A$ between displacements or step increments in the
FIG. 3: Sample trajectories at \( f=4 \text{hz} \) (left) and \( 6 \text{hz} \) (right).

cartesian directions. We use the following definition,

\[
A(\Delta) = \frac{\mathbf{E}[x_i(t)x_i(t+\Delta)]}{\sqrt{\text{Var}[x_i(t)]\text{Var}[x_i(t+\Delta)]}}
\]  

(4)

This is ensemble-averaged to obtain \( \langle A \rangle \) shown in Figure 4. We notice dominant harmonic and sub-harmonic modes at both the pulsing frequencies. The decay of \( \langle A \rangle \) for the lateral transport of particles is predominant compared to their streamwise component at \( f=4 \text{hz} \), while the behavior is similar in the two coordinate directions at \( f=6 \text{hz} \). This is associated with redistribution of energy between harmonic and sub-harmonic modes as explained using Proper Orthogonal Decomposition [12]. The long-range correlation (memory effects) observed in the collective behavior of particles follows the idea of Kac [34], wherein determinism evolves in multi-particle systems governed by stochastic differential equations, which in this case appear to vary depending on the spatial location of particles.

We then examine the behavior of mean-squared displacement (MSD), typically used to study diffusion processes. Ergodicity in PFB is yet to be confirmed, and we use two different measures of MSD. First, is the ensemble averaged MSD given by,

\[
\langle x^2(\Delta) \rangle = \frac{1}{N} \sum_{i=1}^{N} |x_i(\Delta) - x_i(0)|^2
\]

(5)
Second, is the time-averaged MSD given by,

$$\langle \delta^2(\Delta) \rangle = \frac{1}{T - \Delta} \int_0^{T - \Delta} \langle |x_i(t + \Delta) - x_i(t)|^2 \rangle dt$$  \hspace{1cm} (6)$$

which involves both time-averaging and ensemble-averaging operators. $\Delta$ and $T$ refer to lag time and total measurement time. Figure 5 represents MSDs evaluated from particle tracks. Plots of $\langle x^2 \rangle$ confirm that a single value of $\gamma$ cannot be used to describe the anomalous diffusion process. Besides, we notice difference in scaling between $\langle x^2 \rangle$ and $\langle \delta^2 \rangle$ indicating weak non-ergodicity [35–38]. This eliminates the possibility of diffusion in PFB describing transport on a fractal support, which is an ergodic process. We also observe that $\langle \delta^2 \rangle$ approaches a plateau as $\Delta \rightarrow T$ ($\langle x^2 \rangle \sim \Delta^0$) more so at $f=6hz$, reported previously for confined GDPs and SBMs by Metzler et al. [27, 38]. This is in contrast to purely sub-diffusive CTRWs, where plateaus are not present for time-averaged MSD. $\langle x^2 \rangle$ and $\langle \delta^2 \rangle$ show the same limiting behavior at $\Delta/T \rightarrow 0$ and $\Delta/T \rightarrow 1$. The latter is apparent from

FIG. 4: Autocorrelation of x and y displacements at f=4hz (top) and 6hz (bottom).
Equation 6, which has a singularity in the limit of $\Delta \to T$, thus placing the constraint, $\langle x^2 \rangle = \langle \delta^2 \rangle$. Also, $\langle \delta^2 \rangle$ varies $\propto \Delta^1$ over an appreciable time frame for both the values of $f$, previously observed for sub- and super-diffusive unconfined HDPs [38]. This might lead to a false impression of normal Brownian motion unless properly addressed using complementary statistical measures.

To elucidate non-ergodic dynamics in PFB, we define ergodicity breaking parameter (EB) as,

$$\text{EB}(\Delta) = \frac{\langle \delta^2(\Delta) \rangle^2}{\langle \delta^2(\Delta) \rangle^2} - 1$$

(7)

EB quantifies dispersion of time-averaged MSDs, $\delta^2$. We use variation in EB as a function of measurement time, T to describe non-ergodicity, where $\Delta = 0.017s$ is chosen for demonstration. EB for a Brownian motion follows $\lim_{\Delta/T \to 0} \text{EB}_{BM}(\Delta) = \frac{4 \Delta}{3 T}$, indicated by the curve $\propto T^{-1}$ in Figure 6. We observe a more gradual change in EB leading to an asymptotic finite value in the limit of $T \to \infty$. This has been reported previously for anomalous stochastic processes governed by HDPs and CTRWs [39]. To further investigate the nature of ergodicity breaking, we use alternative ergodic parameter, $\mathcal{EB}$ following the definition of Godec and Metzler [40] given by,

$$\mathcal{EB}(\Delta) = \frac{\langle \delta^2(\Delta) \rangle}{\langle x^2(\Delta) \rangle}$$

(8)

A necessary condition for ergodicity is $\mathcal{EB} = 1$, while a sufficient condition is $\text{EB} \to 0$ as $\Delta/T \to \infty$.

In addition, we examine the spread in $\delta^2$ in Figure 7 using the non-dimensional parameter $\xi$ defined as,

$$\xi = \frac{\delta^2(\Delta)}{\langle \delta^2(\Delta) \rangle}$$

(9)

For a Brownian diffusion process, the distribution is Gaussian having a sharp peak around 1 ($\xi = 1$ represents ergodicity). For a smaller value of $\Delta$ (10 time steps), the distribution shows a distinct peak at $\xi > 1$ and $\xi < 1$ at $f=4hz$ and $6hz$. We notice a significant scatter in $\xi$ for growing lag times deviating from ergodic dynamics. $\phi(\xi)$ is finite at $\xi=0$ for all values of $\Delta$, which is true for sub-diffusive CTRWs [41]. The contribution from immobile
FIG. 5: MSD from experiments at $f=4\,\text{hz}$ (top) and $6\,\text{hz}$ (bottom). Dashed lines represent $\Delta^1$ scaling.

Trajectories is more prominent for longer lag times when $f=4\,\text{hz}$. These findings corroborate combination of parent stochastic processes dictating the underlying anomalous diffusion.

Finally, we look at ageing characteristics, which along with ergodicity breaking determine the (non-)stationary nature of a stochastic process. A plot of $\langle \delta^2 \rangle$ as a function of
FIG. 6: Top panel shows EB at f=4hz and 6hz using Δ=0.017s. Dashed lines represent different slopes for guidance. The bottom panel shows variation in $\mathcal{E}B$, reaching an asymptotic value of 1 in the limit of $\Delta \to T$. 
FIG. 7: Histograms of $\xi$ from experiments at $f=4\text{hz}$ (top) and $6\text{hz}$ (bottom).

measurement time, $T$ (Figure 8) reveals a monotonic drop for different values of $\Delta$. This is indicative of a collective sub-diffusive anomalous process. Analogous ageing behavior in a sub-diffusive environment is found in several other systems including plasma membranes as demonstrated by Weigel et al. The apparent diffusivity of the ensemble of particles
decreases as the system evolves longer, but individual behavior could vary depending on the localized states.

FIG. 8: Plots of $\langle \delta^2 \rangle$ as a function of measurement window, $T$ at $f=4\text{hz}$ (top) and $6\text{hz}$ (bottom).

We further use ageing factor Metzler et al. \cite{38} defined as,

$$\Lambda(t_a, \Delta) = \frac{\langle \delta_a^2(\Delta) \rangle}{\langle \delta^2(\Delta) \rangle}$$ (10)

where $\delta_a^2$ refers to the time-averaged MSD considering the ageing time $t_a$ given by,
The above expression is ensemble averaged while calculating $\Lambda$ for different values of ageing time shown in Figure 9. There is a steady drop in $\Lambda$ even for a very small value of $\Delta$. This is a consequence of continued localization of particles in static and quasi-static regions, again indicative of sub-diffusive behavior. Even though the meso-scale response of the system (based on the bubbling pattern) is completely different at $f=4\text{hz}$ and $6\text{hz}$, they exhibit similar anomalous diffusion characteristics.

![Graph](image)

FIG. 9: Plots of $\Lambda$ as a function of dimensionless ageing time, $t_a/T$ at $f=4\text{hz}$ (top) and $6\text{hz}$ (bottom) for different values of $\Delta$ as indicated.
IV. CONCLUSION

We analyzed the anomalous diffusion process having a spatio-temporal dependence in PFB using single-particle tracking, and find traits from a combination of parent stochastic processes. PFB represents a weakly confined multi-particle system having complex non-linear interactions. Finite memory or long-range correlations stem from individual stochastic motions, in line with the ideas of Kac surrounding propagation of chaos [34]. Time-averaged and ensemble averaged MSDs deviate indicating weak ergodicity breaking. MSDs approach a plateau similar to constrained systems for GDPs or SBMs, although the streamwise transport in PFB is effected by the balance between inter-phase drag and gravity. The distribution of amplitude scatter is wide, non-Gaussian, asymmetric, and has a finite contribution at zero stemming from immobile particles, a feature prevalent in CTRWs. The system also exhibits ageing characteristics, which along with ergodicity breaking are suggestive of a non-stationary process. The ageing factor decreases monotonically suggesting an overall subdiffusive process at both the pulsing frequencies, which exhibit different meso-scale behavior altogether. We expect structured flow patterns while lowering the effective diffusivity compared to fluidized systems having an unperturbed inflow, which needs to be verified.

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