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Deterministic separation of suspended particles in a reconfigurable obstacle array

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Abstract

We use a macromodel of a flow-driven deterministic lateral displacement (DLD) microfluidic system to investigate conditions leading to size-separation of suspended particles. This model system can be easily reconfigured to establish an arbitrary forcing angle, i.e. the orientation between the average flow field and the square array of cylindrical posts that constitute the stationary phase. We also consider posts of different diameters, while maintaining a constant gap between them, to investigate the effect of obstacle size on separation. In all cases, we observe the presence of a locked mode at small forcing angles, in which particles move along a principal direction in the lattice. A locked-to-zigzag mode transition takes place when the orientation of the driving force reaches a critical angle. We show that the transition occurs at increasing angles for larger particles, thus enabling particle separation. Moreover, we observe a linear regression between the critical angle and the size of the particles, which allows us to estimate size-resolution in these systems. The presence of such a linear relation would guide the selection of the forcing angle in microfluidic systems, in which the direction of the flow field with respect to the array of obstacles is fixed. Finally, we present a simple model, based on the presence of irreversible interactions between the suspended particles and the obstacles, which describes the observed dependence of the migration angle on the orientation of the average flow.

Introduction

Microfluidic methods that fractionate mixtures into individual chemical or biological components constitute an integral part of micro-total-analysis-systems (μ-TAS). These methods can be broadly classified as active or passive depending on the use or not of an external field to drive the separation. Active methods include dielectrophoresis [1], magnetophoresis [2], acoustophoresis [3], various optical methods [4], [5] and a family of flow field fractionation methods with different fields driving the separative displacement [6]–[9]. Passive methods are generally based on hydrodynamics and particle-solid interactions between the species and the stationary phase in the fluidic system [10], [11]. They include hydrodynamic filtration [12], pinched flow fractionation [13]–[16] and several separation techniques based on
inertial microfluidics [17]. In some cases, hybrid microfluidic separation devices that combine passive and active separation techniques enhance the separation performance [18]–[21]. Deterministic lateral displacement (DLD) is a fractionation technique that can be implemented in all active, passive and hybrid modes. First introduced as a passive method [22], DLD exploits the experimental observation that particles of different size flowing through a periodic array of cylindrical posts may migrate in different directions, thus leading to separation (Figure 1). Recent work suggests that DLD could also effectively fractionate a mixture based on the shape of the particles [23],[24]. The ability to continuously separate particles of different size or shape has made DLD a popular method for the fractionation of a number of biological samples using microfluidics [25]–[31]. Although DLD was initially introduced as a flow-driven, passive microfluidic method, we have shown in previous work that driving the particles by external forces also results in the separation of individual components. Specifically, we have successfully used gravity and electric fields to drive the separation of suspended particles in force-driven DLD (f-DLD) [32]–[35].

In their original work, Inglis et al. observed two types of trajectories, depending on the size of the particles: bump mode trajectories for large particles and zigzag mode trajectories for small ones [36]. A bump mode trajectory is schematically shown by the bigger circles in Figure 1, and corresponds to a situation where the particle moves alongside a line of obstacles on the lattice (a column of obstacles, along solid line C in the figure). A zigzag mode trajectory is schematically shown by the smaller circles in Figure 1, and corresponds to trajectories in which the particle zigzags inside the array. Inglis et al. postulated that zigzag mode trajectories were, on average, aligned with the flow direction. However, Kulrattanararak et al. later reported that particles in zigzag mode do not necessarily move in the direction of the flow [37], [38]. In general, as the size of the particles increases, their motion eventually transitions from zigzag mode to bump mode for particles larger than a critical size, a transition that depends on the geometry of the lattice and the orientation of the average flow. In previous work, we have shown that f-DLD results in similar behavior [39]. Moreover, using scaled-up macromodels of microfluidic DLD systems, we were able to investigate the entire

![Schematic view of a DLD separation system. The large (small) solid circles represent the position of a large (small) particle at increasing times. The open circles represent the cylindrical obstacles. The solid line L denotes the direction of particle migration, the arrow F represents the direction of the flow and the solid line C connects centers of obstacles aligned in a lattice column. The dashed line L’ that is parallel to L is drawn to better illustrate the migration angle (β).](image)
range of possible orientations of the external force with respect to the array of obstacles, and show that, in fact, all particles transition from bump mode to zigzag mode as the forcing angle increases [32], [39], [40]. The forcing angle $\alpha$ is the angle between a column of obstacles in the lattice and the average flow (or external force), and the migration angle $\beta$ is the angle at which the particles migrate on average, also measured with respect to a column in the array (see Figure 1). The macromodel experiments showed that, as the forcing angle increases from zero, all the particles remain locked to move alongside a column of obstacles in the array, i.e. $\beta=0^\circ$, until the forcing angle reaches a critical value, $\alpha_c$, which depends on particle size. Then, for each particle size, when the forcing angle is larger than their critical angle, the particles are able to move across columns of obstacles, resulting in a periodic zigzag motion, with $\alpha \neq \beta$ in general. In fact, for any forcing angle, the motion of the particles is periodic and the average migration is always along a lattice direction. A simple geometric model discussed below captures the observed dynamics by considering the effect of non-hydrodynamic interactions between suspended particles and solid obstacles [32], [39], [41].

Here, we use macromodels of flow-driven DLD devices to investigate conditions leading to size-separation of suspended particles, depending on the geometry of the array of obstacles and the average orientation of the pressure-driven flow. Specifically, we consider two arrays with different size of cylindrical posts, and continuously vary the direction of average flow to cover a wide range of forcing angles ($0 < \alpha < 30^\circ$).

In contrast, the forcing angle in previous experiments investigating flow-driven DLD microfluidic devices is built into the system and could not be modified. As a result, such devices were only able to fractionate a sample between particles larger and smaller than a critical size that is fixed by the geometry of the system. The present set of experiments shows that the motion of suspended particles is analogous to that observed in the f-DLD case. Specifically, we experimentally show: (i) the existence of a locked mode for all particle sizes, in which the average migration angle is $\beta=0^\circ$ (particles move along a column of obstacles) for a range of forcing angles $\alpha_c > \alpha > 0$; (ii) a sharp transition from locked mode to zigzag mode in which particles move periodically at certain lattice directions; (iii) a monotonic increase in the critical angle at which the locked-to-zigzag transition occurs with particle size. Finally we present a simple model, based on the irreversible nature of non-hydrodynamic interactions between the suspended particles and the obstacles, which accurately describes the migration angle over the entire range of forcing angles.

### Experimental Setup

Our experimental setup is a scaled-up version of a microfluidic DLD system, consisting of a closed channel of width $L = 280\text{mm}$ fabricated using acrylic plates (see Figure 2A). A square array of obstacles ($l = 200\text{mm}$) is centered in the channel
and a circular disk is cut at the center of the array as shown in Figure 2B. The rotating central part allows us to vary the forcing angle continuously. Additionally, to study the effect of obstacle size on particle trajectory, two different arrays are used. We show a schematic of the geometry of the arrays in Figure 2C. The difference between the two arrays is the diameter of the obstacles, either \( d = 1 \text{mm} \) or \( d = 2 \text{mm} \). The height of the channel (and obstacles) is \( h = 5 \text{mm} \) and the open gap between obstacles is \( 4 \text{mm} \), in both arrays. They were fabricated using a 3D printer (Objet350 Connex, Stratasys). The reason for the rectangular shape of the complete array is to ensure a uniform flow over the width of the channel (except close to the walls). A circular array alone, in contrast, would not provide a uniform flow resistance over the width of the channel and could lead to significant flow variations. In addition, the Hele-Shaw type of configuration used in the experiment, in which the length of the channel is much larger than its height, \( l \gg h \), makes entrance effect negligible for low Reynolds number flows. The flow is driven by a constant pressure drop generated by a Mariotte’s bottle and distributed over the channel width using a manifold at the inlet.

In order to compare our results with microfluidic systems we have to satisfy both geometric and dynamic similarity. To this end, we maintain the shape of the posts (cylindrical), the spacing between the obstacles relative to their size, and the particle/obstacle aspect ratios comparable to those used in microfluidic systems, thus satisfying geometric similarity. In order to establish dynamic similarity, we need to maintain relatively low Reynolds numbers. Therefore, we use a mixture of glycerin (99% Glycerin, McMaster-Carr) and water with a volume ratio 3:2. The dynamic and kinematic viscosity of the liquid mixture is approximately \( \mu = 0.016 \text{Ns/m} \) and \( \nu = 1.38 \times 10^{-5} \text{ m}^2/\text{s} \), respectively [42]. The flow rate in the experiments is approximately \( Q = 8 \text{ cm}^3/\text{s} \), and the corresponding Reynolds number can be estimated as \( Re = Qh/Av \approx 3 \), where \( A = l \times h = 10 \text{cm}^2 \) is the cross section area of the channel. We note however, that our goal is not to reach the Stokes limit of vanishingly small Reynolds numbers but to ensure dynamic similarity between our macromodel and microfluidic systems. In fact, previous work in microfluidics has shown that DLD remains an effective separation method at moderate Reynolds numbers, e. g. \( Re \lesssim 40 \) [43], [44]. In terms of Brownian motion, microfluidic DLD devices work in the deterministic regime, \( Pe \gg 1 \), a condition clearly satisfied in macroscale models. Finally, we make sure that the Stokes numbers (St) in the macromodel experiments are also small. We used nylon particles (density \( \rho = 1.13 \text{g/cm}^3 \)) of six different sizes, with diameters \( D = 1/16'' \) (1.59 mm), \( 3/32'' \) (2.38 mm), \( 1/8'' \) (3.18 mm) (McMaster-Carr), \( 0.072'' \) (1.83 mm), \( 7/64'' \) (2.78 mm) and \( 9/64'' \) (3.57 mm) (Precision Plastic Ball Co.). We estimate a particles Reynolds number \( Re_p = \frac{u_p D}{\nu} \), where \( u_p \) is the velocity of the particles, between 0.3 and 1.85,
depending on particle size. Therefore, the corresponding Stokes numbers, \( \text{St} = \frac{1}{9} \left( \frac{\rho_p}{\rho_l} \right) Re_p \), are in fact small, between 0.03 and 0.2.

Figure 2 a) Schematic view of the experimental setup. b) Top and side view of the array of obstacles. c) Representation of the geometry of the array.

Independent experiments are performed for each particle size, obstacle array and forcing angle. In each experiment, we analyze the trajectory of \( \sim 20-30 \) particles and determine the average migration angle.

Results and Discussion

First, we investigate the existence of a locked mode, in which particles move along a column of obstacles (\( \beta = 0^\circ \)) for forcing angles lower than a certain critical angle. Then, we characterize the transition into the zigzag mode as the forcing angle increases beyond the critical value. To this end, we introduce the probability of crossing \( P_c \), defined as the fraction of particles that move in zigzag mode over the total number of particles analyzed. Alternatively, \((1 - P_c)\) is the fraction of particles locked to move in the \([1, 0]\) lattice direction without crossing columns of obstacles (\( \beta = 0^\circ \)). The results are presented in Figure 3. Clearly, in all cases, we observe a sharp transition from no crossing (i.e. locked mode at \( \beta = 0^\circ \)) to complete crossing with \( P_c = 1 \). Therefore, we define the critical angle for each particle as the forcing angle at which the crossing probability is \( P_c = 1/2 \) (calculated by linear interpolation using the closest data points with probabilities higher and lower than 1/2). It is also clear in Figure 3 that the locked-to-zigzag transition occurs at increasing forcing angles for particles of increasing size, which demonstrates that particles can be separated by size. In addition, the fact that we observe sharp transitions without overlap between different curves indicates that these particles could be separated with high resolution.
Figure 3 a) Probability of crossing for different size of particles for the array with smaller obstacles (1 mm obstacle diameter). b) Probability of crossing for different size of particles for the array with larger obstacles (2 mm obstacle diameter).

In Figure 4, we present the critical angle as a function of particle size for the two different arrays of obstacles. Interestingly, we observe a linear relationship for both
arrays. In contrast, the model proposed by Inglis and coworkers, based on the streamlines in the absence of particles (streamlines model), predicts a cubic relation (solid line in Figure 4). We note that, in the case of large particles (e.g., $D > 3$ mm), inertia effects might contribute to the discrepancy between the experimental results and the streamlines model. On the other hand, we also observe deviations from the streamlines model with small particles (e.g., $D < 3$ mm), in which case inertia effects are probably negligible, as indicated by the small Reynolds (and Stokes) numbers. Finally, we also note that the suspended particles are not completely neutrally buoyant, but slightly lighter than the suspending fluid, and would therefore migrate closer to the top wall of the channel. This could also contribute to the difference with the predictions of the streamlines-based model, which does not consider changes in the in-plane velocity depending on the vertical position of the particles.

Figure 4 Critical angles as a function of particle size in both arrays. Particle diameter is nondimensionalized by the gap size. Straight lines correspond to a linear fit of the results. The solid curve is calculated using the model proposed by Inglis et al. [36]. In the array with smaller obstacles, the linear fit is $\alpha_c = 36.2861(D/g) - 6.90216, R^2 = 0.9986$. In the array with bigger obstacles, the linear fit is $\alpha_c = 28.7769(D/g) - 2.81352, R^2 = 0.9956$.

The linear dependence of the critical angle on the size of the particles allows us to estimate the size resolution that is possible with the corresponding separation system. Specifically, we first estimate the largest uncertainty in the determination of the critical angle by the largest variance (error bar) reported in Figure 4, $\delta \rho \sim 2^\circ$. Then, we estimate the corresponding uncertainty in the radius of the particles using the
linear correlation, and obtain $\Delta \alpha \lesssim 150 \mu m$ ($\Delta \alpha \sim 140 \mu m$ and $\Delta \alpha \sim 110 \mu m$ for the arrays with 1mm and 2mm obstacles, respectively). Therefore, we expect excellent separation for particles that have a difference in size $\Delta \alpha \gtrsim 300 \mu m$. For example, let’s consider a binary mixture of particles with radius $a_1 = 0.9 \ mm$ ($D_1 = 1.83 \ mm$) and radius $a_2 = 1.2 \ mm$ ($D_2 = 2.38 \ mm$), that is approximately $300 \mu m$ difference in radius. Then, using the lattice with 2 mm obstacles and a forcing angle $\alpha \approx 12^\circ$ would result in zero crossing for the large particles and complete crossing for the small ones, corresponding to ideal purity in the separation.

Figure 5 a) Migration angle as a function of forcing angle (1 mm diameter obstacle). b) Migration angle as a function of forcing angle (2 mm diameter obstacle). The dashed line represents $\beta = \alpha$ (in both plots).
Second, we investigate the migration angle in the zigzag mode. As we discussed in the introduction, the original DLD work by Inglis and coworkers assumed that in zigzag mode particles move, on average, parallel to the forcing angle. However, further analysis showed that this is not the case [37], [38]. Similarly, our f-DLD experiments clearly indicated that particles move periodically at specific lattice directions that, in general, are not aligned with the external force [32], [39]. In Figure 5, we present the migration angle as a function of the forcing angle. Although in most cases the migration angles are similar to the forcing angle, there are some cases in which it is clear that $\beta \neq \alpha$.

**Migration Model**

To explain our experimental results, we propose a model based on the individual particle-obstacle interactions that take place as the suspension moves through the array of obstacles. We refer to the motion of a suspended particle around and past a cylindrical post as a particle-obstacle collision. We assume that during such a collision particles may experience irreversible interactions that lead to a net lateral displacement. (Note that these assumptions, as well as the resulting model, are completely analogous to those used to describe f-DLD systems, and a more detailed discussion can be found elsewhere [39], [41], [45].) In fact, collisions can be divided into two groups depending on the initial offset, as schematically shown in Figure 6. Collisions for which the initial offset, $b_{in}$, is larger than a certain critical value $b_c$, $b_{in} > b_c$, are reversible and there is no net lateral displacement resulting from the collision. In this case, trajectories are fore-and-aft symmetric. On the other hand, collisions for which the initial offset is small, with $b_{in} < b_c$ (see shaded region in Figure 6), are irreversible and their outgoing offset is $b_c$. In other words, irreversible collisions result in a net lateral displacement of magnitude $(b_c - b_{in})$. Note that the resulting collapse of irreversible trajectories, according to which any incoming particle with $b_{in} < b_c$, i.e. inside the shaded region in Figure 6, comes out of the collision with the same offset $b_c$, implies the existence of directional locking. That is, only some migration angles are possible and each migration angle is associated with a range of forcing angles. There is no one-to-one correspondence between $\alpha$ and $\beta$, on the contrary, each migration angle is associated to a finite range of forcing angles that lead to it [40], [41], [46].
Given particle size and geometry of the obstacle array, the critical offset is the only unknown parameter in the proposed collision model. It can be calculated from the critical angle using the relation $b_c = d \sin(\alpha_c)$ as shown in Figure 6 [41]. After the critical offset is determined, it is straightforward to calculate the migration angle as a function of the forcing angle from geometric considerations, given that the result of every particle-obstacle collision can be predicted. In fact, only those collisions that are irreversible need to be accounted for and they simply result in a net lateral displacement perpendicular to the forcing direction [41], [46]. In Figure 7, we show the comparison between the proposed model and the experimental results. Note that, the critical offset is determined from the crossing probability. Therefore, the comparison between the model and the migration angles at forcing angles larger than the critical one has no fitting parameters. In general, we observe good agreement between the model and the experimental results, which suggests that in the zigzag mode the migration angle is not necessarily the same as the forcing angle but rather results from directional locking into certain lattice directions.
Figure 7 Comparison of experimental results with the proposed collision model for particles of all sizes and in both arrays. In each graph, the solid (dot dashed) line corresponds to the results predicted by our model in the lattice with small (large) obstacles. The critical offset was calculated from the crossing probability. Solid diamonds (squares) correspond to the experimental results obtained in the lattice with small (large) cylinders.

Finally, we obtain a second estimate of the critical angle for each combination of particle and obstacle size, by directly fitting the measured migration angles. Specifically, we fit the migration angles using the collision model and $b_c$ as a fitting parameter. The results are nondimensionalized by the obstacle radius and presented in Figure 8. We compare both sets of critical angles for each array of obstacles, one set of values calculated from the critical angles (estimated from the crossing probability measured experimentally) and one set of values obtained from the fitting of the migration angles. Note that, in most cases, due to the stair-case type of curve predicted by the model, the fit of the migration angles results in a range of critical offsets instead of a single value. Therefore, in Figure 8, we report the average value and the error bars correspond to the uncertainty interval. We observe, in both arrays, good agreement between the two sets of critical offsets. Interestingly, we additionally observe that the nondimensional critical offset follow a universal linear trend as a function of the nondimensional particle size. The results need to be further validated
at the micro scale, but this simple correlation could provide the necessary information to tailor the design of DLD systems to specific applications.

Figure 8 Nondimensional critical offset as a function of nondimensional particle size. The circle (square) symbols correspond to the array with large (small) obstacles. The two sets of $b_c$ values correspond to the calculation based on the crossing probability (solid symbol) or the best fit of the migration angle with the collision model (open symbol). The solid line is a linear fit of both sets of critical offsets ($b_c/R = 0.66674 D/R − 0.34982, R^2 = 0.94196$).

Conclusions

We used a macromodel to investigate flow-driven DLD microfluidic systems over a wide range of forcing angles (direction of the average flow with respect to a column in the array of obstacles). We demonstrated the existence of a locked mode for all the different particles considered here and in two arrays with different obstacle sizes. In this locked mode, corresponding to small forcing angles, the migration angle of the particles remains $\beta = 0^\circ$ until a sudden transition into zigzag mode occurs when the forcing angle reaches a critical transition angle. The fact that the transition occurs at increasing forcing angles for larger particles enables particle separation. In fact, we observed a linear trend for the critical angle as a function of particle size. We note that this experimental finding disagrees with the model originally proposed by Inglis and coworkers [36]. In addition, we showed that a simple collision model, based on
irreversible particle-obstacle interactions, not only captures the sharp *locked-to-zigzag mode* transitions, but also predicts the migration angles at larger forcing angles. Unfortunately, the prevalent DLD experiments in microfluidics have been focused on small and fixed orientations of the driving flow field and no general results are available for the behavior of particles of different size as the forcing angle increases. Therefore, further microfluidic experiments are needed to validate the linear trend in the critical angle as a function of particle size observed in the macromodels used here.

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