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# ACCURACY OF EXTERNAL FORCE MEASUREMENTS BASED ON PARTICLE TRACKING VELOCIMETRY

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#### ABSTRACT

In microfluidic systems external forces are frequently applied to fluids or colloidal suspensions in order to accomplish or enhance mass transport tasks. The complexities of microscale geometries and material properties, however, can cause discrepancies between theoretical predictions and the actual values of the applied force. Therefore a calibration experiment is necessary to validate the actual magnitude of the applied force. One method of such in vivo calibration is through observations of tracer particle motions using particle tracking velocimetry (PTV). In microfluidic applications, the tracer particles of choice are typically submicron in diameter and therefore undergo significant Brownian motion. Further complicating the matter is the presence of the solid channel boundaries whose presence can lead to hindered Brownian motion and position-dependent hydrodynamic drag. In this paper we present a Langevin simulation study of the effects of normal and hindered Brownian motions, and the time between image acquisitions on the accuracy of external force measurements based on PTV. It is found that the relative strength between the random forces that cause Brownian motion and the applied external force plays a critical role in measurement accuracy. We also found that hindered Brownian motion and the associated sampling trajectory biases contribute additional force measurement inaccuracies when PTV is conducted in the vicinity of a solid boundary.

## 1. INTRODUCTION

In many micro- and nanofluidic applications, it is frequently desirable to transport fluid, molecules and colloidal particles to accomplish tasks such as mixing, drug delivery, adhesion promotion, and DNA analysis. In these applications, ensemble transport beyond the random Brownian motion and diffusion is usually achieved by applying long range conservative forces. Examples of such forces include electrostatic attraction and repulsion [1], electroosmosis [2], electrophoresis [3] and dielectrophoresis [4], magnetic manipulation [5-7], optical scattering force [8], and optical gradient force [9]. In most cases, these mechanisms apply an additional linear force to individual particles to increase their displacement within a limited amount of time. While one can calculate the magnitude of the applied "push" based on established theories, local variations can still exist due to complex geometry, local fluid properties and particle variability. Thus, it is important to be able to calibrate and measure the magnitudes of the applied forces *in vivo* and to confirm that the behaviors of these particles conform to that of theoretical predictions.

One possible method to accomplish such *in vivo* force calibration measurement is through particle tracking velocimetry (PTV). Briefly, PTV follows the path of individual tracer particles in fluids through sequential imaging under a microscope [10, 11]. By conducting particle identification and locating particle centers with subpixel resolutions, the center positions of an ensemble of particles can be obtained as a function of time. Subsequently, time- and ensemble-averaged motions of the tracer particles can be obtained and the magnitude of the applied force can be extracted. Schmidt *et al.* [8] has recently reported using PTV to estimate the evanescent wave scattering force applied on polystyrene microspheres transported along an optical waveguide.

While PTV is an attractive and convenient method to conduct fluidic and colloidal measurements in the micro- and nanoscales, recent reports by Huang *et al.* [12] and Sadr *et al.* [13] have indicated that Brownian motion of tracer particles can cause significant measurement bias. These researchers also found that the accuracy of PTV depends on the size of the particles as well as the time elapsed between consecutive image acquisitions. Furthermore, particles experience anisotropic increase in mobility and hindered Brownian motion when approaching other particles or flow channel boundary. These boundary-induced hydrodynamic drags on particles can further lead to additional inaccuracy of PTV measurements. In this paper, we extend the Langevin simulation method of Huang *et al.* [12] to computationally simulate PTV measurement of applied external forces on an ensemble of isolated colloidal particles and examine the effect of Brownian motion on force measurement accuracy.

#### 2. THEORY

The proper examination of particle-based velocimetry accuracy by means of numerical simulations requires a fundamental and thorough understanding of the dynamic theories of near-wall tracer particles. In particular, the motions of these particles, as depicted in this simulation, are affected by both hindered and unhindered Brownian motion. In the present section we introduce the Langevin equation for a near-wall particle and effectively incorporate necessary dynamic forces to ascertain a non-dimensional equation of motion that will provide the foundation for our numerical simulation.

## 2.1. The Langevin Equation

In a Langevin simulation, particle displacements are computed based on a stochastic equation [14]. The simulation geometry is depicted in figure 1. A neutrally buoyant tracer sphere of radius *a* is free to undergo two-dimensional Brownian motion in the vicinity of a solid/fluid boundary. In addition, the particle is under the influence of a constant external force  $f_x$  in the direction parallel to the solid boundary. The displacement of a particle experiencing the aforementioned conditions between time  $t_i$  and  $t_{i+1}$  is given as,

$$x_{i+1} - x_i = \frac{dD_x}{dx}\delta t + \frac{D_x}{k_B\Theta}f_x\delta t + N(0,\sqrt{2D_x\delta t}) \quad (2.1.1)$$

$$z_{i+1} - z_i = \frac{dD_z}{dz}\delta t + \frac{D_z}{k_B\theta}f_z\delta t + N(0,\sqrt{2D_z\delta t}) \quad (2.1.2)$$

where time interval  $\delta t = t_{i+1} - t_i$ . Regarding the above equations, (x,z) is the particle's center position,  $k_B$  is the Boltzmann constant, and  $\Theta$  is the temperature of the fluid.  $D_x$  and  $D_z$  are the diffusion coefficients parallel and normal to the wall, respectively. It should be noted that at distances far away from the wall (z >> a), where particle-wall interaction is virtually non-existent,  $D_x$  and  $D_z$  are both equal to the Stokes-Einstein diffusivity,  $D_0 = k_B \Theta/6\pi\eta a$ , where  $\eta$  is the fluid viscosity. Due to the fact that near-wall particle motion is anisotropic and hindered,  $D_x \neq D_z$ , and both are less than or equal to  $D_0$ . Furthermore,  $D_x$  is a function of z alone [15], and consequently its spatial derivative,  $\frac{dD_x}{dx} = 0$ , in the above equation. A



Figure 1. A schematics of simulation geometry.

discussion concerning the z-dependency of  $D_x$  and  $D_z$  will be postponed for later sections.  $f_x$  and  $f_z$  represent the external forces acting on the particle in the x - and z -directions, respectively. A density mismatch between the particle and the surrounding fluid can potentially give way to a buoyancy force in the vertical direction. For monodispersed, particles with a density greater than of the surrounding fluid, this buoyancy force is equivalent to

$$f_z = f_g = \frac{4\pi}{3} a^3 \Delta \rho g \tag{2.1.3}$$

where  $\Delta \rho$  is the difference in density between the particle and surrounding fluid and g is the gravitational acceleration. Because most near-wall velocimetry measurements utilize density-matched tracer particles,  $f_z$  is much less significant than the thermal energy-driven Brownian motion and therefore assumed to be negligible. The last term in both equations (2.1.1) and (2.1.2),  $N(0, \sqrt{2D_x \delta t})$ , denotes hindered Brownian motion in the form of stochastic particle displacements randomly sampled from a normal distribution with a zero mean and standard deviation of  $\sqrt{2D_{x,z}\delta t}$ .

Equations (2.1.1) and (2.1.2) are non-dimensionalized by using particle radius, a, as the length scale and  $a^2/D_0$  (the time required for an isolated particle to travel a distance of one radius due to its unhindered Brownian motion) as the time scale. By utilizing these two scaling parameters, the equations of motion can be transformed into

$$X_{i+1} = X_i + F_x \beta_x \delta T + N \left( 0, \sqrt{2\beta_x(Z_i)\delta T} \right) \quad (2.1.4)$$

$$Z_{i+1} = Z_i + \frac{d\beta_z}{dZ}\Big|_{Z_i} \delta T + N\left(0, \sqrt{2\beta_z(Z_i)\delta T}\right) \quad (2.1.5)$$

where  $X \equiv x/a$ ,  $Z \equiv z/a$ , and  $T \equiv D_0 t/a^2$  are the non-dimensional x, z, and t, respectively. The magnitude of the external force is characterized by  $F_x = f_x a/k_B \Theta$ , its relative strength to the thermal-energy driven random force. Thus the motion of the particle is dictated by both Brownian motion and external forcing. Finally, hindered Brownian motions in both x and z directions are given by

$$\beta_x \equiv \frac{D_x(z)}{D_0}, \quad \beta_z \equiv \frac{D_z(z)}{D_0}$$
(2.1.6)

#### 2.2. Hindered Brownian motion

Hydrodynamic effects cause the near-wall tracer particles to undergo anisotropic hindered Brownian motion. According to Goldman *et al.* [15], the hindered diffusion coefficient can be approximated by the following equation,

$$\frac{D_x}{D_0} \equiv \beta_x(Z) = 1 - \frac{9}{16}(Z)^{-1} + \frac{1}{8}(Z)^{-3} - \frac{45}{256}(Z)^{-4} - \frac{1}{16}(Z)^{-5} + O(Z)^{-6}$$

which is more accurate for Z > 2 ("Method of Reflection"). Goldman *et al.* [16] also proposed an approximation through the following asymptotic solution for Z < 2:

$$\frac{D_x}{D_0} \equiv \beta_x(Z) = -\frac{2[\ln(Z-1) - 0.9543]}{[\ln(Z-1)]^2 - 4.325\ln(Z-1) + 1.595]}$$

Bevan & Prieve [17] derived a simplified diffusion constant,  $D_Z$ , for near-wall particle motions in the direction normal to the solid boundary:

$$\beta_{z}(Z) = \frac{6 (Z-1)^{2} + 2 (Z-1)}{6 (Z-1)^{2} + 9 (Z-1) + 2}$$

Because these hindered diffusion coefficients are dependent on Z, distance between the particle under consideration and the solid boundary, they must be updated at every time step during the Langevin simulation to ensure accuracy.

# 3. IMPLEMENTATION OF LANGEVIN SIMULATIONS

Our Langevin simulations were conducted under the assumption of no particle-particle interaction due to the fact that most velocimetry experiments are carried out using dilute particle suspensions. At the beginning of each simulation, the position of a particle center is positioned at (0, Z), where Z is randomly chosen in the range of 1 < Z < 100. Once initiated, the simulation advanced for a total of  $\Delta T/\delta t$  steps while updating  $\beta_x$  and  $\beta_z$  and recording the position of the particle (X,Z) after each step. With the solid boundary located at Z = 0and the nondimensional radius of the particle being unity, the smallest Z value a particle could have was Z = 1, implying contact with the wall. A boundary condition was needed in the simulation to prevent a particle from entering the solid wall during a simulation step. The studies conducted by Peters & Barenbrug [18, 19] concerning the efficiencies of different boundary conditions for Langevin simulations influenced us to use a simple yet effective specular reflection to avert particles from entering the solid wall. Because the particle has no limitations regarding its movement away from the wall (which is governed by Brownian motion), specular reflection is the only boundary condition applied in our simulation.

The single particle simulation was then repeated  $10^5$  times to obtain a large statistical sample. In order to ensure that the results were a consequence of physical parameters only, the random number generator, which controls the initial positions of the particles, was seeded identically for all simulation trials.

Lastly, the choice of the computational time step size,  $\delta T$ , is influenced by two physical constraints [14]. First, the time step must be greater than the particle momentum relaxation time,  $mD_0/k_B\Theta$ , where m is the mass of one particle. The nondimensional form of this is given as,

$$\delta T \gg \frac{m D_0^2}{k_B \Theta a^2} \sim O(10^{-6})$$
 (3.1)

for a > 100 nm. Secondly, numerical accuracy requires that the time step must be short enough such that the diffusion coefficients, their gradients, and particle hindered mobility are essentially constant during the time step. In order to meet these

criteria,  $\delta T$  was chosen to be 10<sup>-4</sup>, which satisfies both constraints and is computationally efficient.

## 4. RESULTS AND DISCUSSIONS

#### 4.1. Definition of Force Measurement Accuracy

We define the accuracy of force measurement by the quantity  $\langle F_x \rangle / F_x$ , where  $\langle \rangle$  represents ensemble average and  $\langle F_x \rangle$  and  $F_x$  are the measured and applied magnitudes of force experienced by the tracer particle ensemble, respectively. By this definition,  $\langle F_x \rangle / F_x = 1$  would suggest a perfect measurement while deviation from unity represents a measure of measurement bias. A simple manipulation of equation (2.1.4) reveals that the magnitude of external force experienced by the particles can be obtained from their average velocity. Mathematically this is equivalent to

$$\langle F_{x} \rangle = \langle \frac{\Delta X - N(0, \sqrt{2\beta_{x}\Delta T})}{\beta_{x}\Delta T} \rangle, \qquad (4.1.1)$$

where  $\Delta X$  is the displacement of a tracer particle during time  $\Delta T$ . Using these definitions, we examined the accuracies of particle-velocimetry-based force measurements due to both unhindered and hindered Brownian motions.

## 4.2. Effects of Brownian Motion on Force Measurement Accuracy

Using our simulation results, we first examine the case of isolated tracer particles where the particles under consideration are very far away from the solid boundary (Z > 50) such that particles are pulled by a constant force  $F_x$  while undergoing unhindered Brownian motion. For these particles, equation (4.1.1) reduces to

$$\langle F_{\chi} \rangle = \langle \frac{\Delta X - N(0, \sqrt{2\beta_{\chi}\Delta T})}{\beta_{\chi}\Delta T} \rangle = \frac{\langle \Delta X \rangle}{\Delta T}.$$
 (4.2.1)

because  $\beta_x = 1$ ,  $\langle N(0, \sqrt{2\beta_x \Delta T}) \rangle = 0$  and  $\Delta T$  is a constant. Simulation results of various external force magnitudes were first analyzed to obtain their respective measurement accuracy



Figure 2. Accuracy of force measurements for various force magnitudes applied to isolated tracer particles undergoing unhindered Brownian motion.

and are shown in figure 2. In figure 2, it can be observed that force measurement accuracy degrades significantly at  $\Delta T < 10^{-1}$  for all applied force magnitudes. The inaccuracy is particularly pronounced at  $F_x < 1$ , where thermal fluctuation force of Brownian motion dominates over the external force. The force measurement accuracy becomes quite reliable for all  $\Delta T$  once  $F_x$  is greater than five.



Figure 3. Average force measured through 20 groups of 5000 particles undergoing unhindered Brownian motion with an external applied force of  $F_x = 1$ .



Figure 4. Standard deviation of the average force measured through 20 groups of 5000 particles undergoing unhindered Brownian motion with an external applied force of  $F_x = 1$ .

The potential 400% measurement inaccuracy at small  $F_x$ and  $\Delta T$  drew our attention to suspect that the probabilistic nature of Brownian motion can lead to significant measurement variations from particle ensemble to ensemble. Figure 3 reveals that significant inaccuracy and trial-to-trial variability occurring at  $\Delta T < 10^{-1}$ , suggesting that force measurement of short interacquisition time can result in large amount of force measurement errors. Further supporting this notion is figure 4, which shows the standard deviation of measured force magnitude by 20 particle groups of same size as a function of  $\Delta T$ . Notice that the standard deviation follows an almost exponential decay and only becomes less than 0.1 at above  $\Delta T$ >  $10^{-1}$ . It is evident that repeatable, accurate force measurements would require both  $\langle \Delta X \rangle / \Delta T = F_x$  and a small



Figure 5. Ratio of the effect of Brownian motion to that of an externally applied force of  $F_x = 1$  on particle displacements.

standard deviation of  $\langle \Delta X \rangle / \Delta T$ . Since Brownian motion is the only stochastic process involved, it is most likely that the measurement inaccuracies and fluctuations are the results of Brownian motion dominance over deterministic external force effect.

A comparison regarding the relative effects of Brownian motion and the applied external force on a group of particles is shown in figure 5 as a ratio between the magnitudes of the mean random molecular force leading to Brownian motion,  $\langle F_{BM} \rangle$ , and the applied external force. Mathematically, this ratio is given as,

$$\frac{\langle F_{BM} \rangle}{F_{\chi}} = \frac{\frac{1}{20} \sum_{i=1}^{20} \sigma_i(\Delta T)}{\alpha(\Delta T)}$$

where  $\sigma_i(\Delta T)$  is the standard deviation of displacements within each particle group and  $\alpha(\Delta T)$  is the displacement a particle would experience under an applied force,  $F_x$ , and constant drag with no Brownian motion. It can be observed in this figure that at  $\Delta T < 10^{-1}$  the influence of Brownian motion on the average motion of a particle ensemble is much more significant than the external force. Under this time scale, Brownian motion severely hinders force measurement accuracy and can achieve magnitudes in excess of 80 times that of the applied force in extremely short time. The random nature of Brownian motion thus explains the severe degree of fluctuation in force measurement accuracy shown in figures 2 to 4 for  $\Delta T < 10^{-1}$ .

Another important feature concerning figure 5 is that the applied external force begins to dominate over Brownian motion in particle displacements at  $\Delta T > 1$ . This also explains the observations made in figures 2 to 4 that high force measurement accuracy can be obtained at  $\Delta T > 1$ . Theoretically, this can be explained by the fact that the effects of the random forces that cause Brownian motion,  $F_{BM}$ , on particles is expressed through the spread of the particle displacements scales with  $\Delta T^{1/2}$ , whereas the effects of the externally applied force,  $F_x$ , produces a uniform displacement and scales with  $\Delta T$ . This physical scaling argument of relative dominance further supports the fact that both  $F_x$  and  $\Delta T$  are critical factors to consider when conducting particle-based force measurements.

#### 4.3. Effects of Near-Wall Hindered Brownian Motion on Force Measurement Accuracy

We subsequently examined the effects of the wall presence and hindered Brownian motion on force measurement accuracy. While equation (4.1.1) still provides the analytical formula for calculating the measured magnitude of the externally applied force, the calculation is much more complicated because  $\beta_x \neq 1$  and is Z-dependent. Still, some manipulation of statistical formula can reduce equation (4.1.1) to

$$\langle F_x \rangle = \langle \frac{\Delta X - N(0, \sqrt{2\beta_x(Z)\Delta T})}{\beta_x(Z)\Delta T} \rangle = \frac{1}{\Delta T} \langle \frac{\Delta X}{\beta_x(Z)} \rangle, \qquad (4.3.1)$$

since  $\langle N(0, \sqrt{2\beta_x\Delta T}) \rangle = 0$  and  $\Delta T$  is a constant. That is, a good measure of the applied force can be obtained if one follows the trajectory of a particle to obtain (X,Z) as a function of time and subsequently compute the ensemble average of the trajectory dependent  $\beta_x(Z)$  and  $\langle \Delta X/\beta_x(Z) \rangle$ . Unfortunately, while it is possible to determine the instantaneous threedimensional position of particle in near-surface particle tracking velocimetry [12, 20, 21], determining the trajectories that particles take between image acquisitions is an impossible task. Therefore, the best one can do is to estimate the value of  $\beta_x$  that a particle experiences during the measurement  $\Delta T$ . A simple way of estimation is to estimate  $\beta_x$  by evaluating its value at  $Z_0$ , the starting Z position of a particle. Therefore, equation (4.3.1) becomes

$$\langle F_x \rangle = \frac{1}{\Delta T} \langle \frac{\Delta X}{\beta_x(Z_0)} \rangle$$
 (4.3.2)

which contains only quantities that can be measured and calculated in a near-wall particle tracking velocimetry experiment.

Figure 6 shows the force measurement accuracy obtained from particles from near-wall regions of various depths. The first observation one can make is that larger sampling depth produces better measurement accuracies for all  $\Delta T$ . Also, for small  $\Delta T$ , particle tracking velocimetry tends to under-estimate the actual magnitude of the external force. Finally for large  $\Delta T$ , measurements of particle displacements very close to wall tend to over-estimate the actual external force, while force measurements obtained from large measurement depth provides much better accuracy. Force measurement accuracies obtained from displacements of tracer particles at various initial distances from the solid boundary are also examined and presented in figure 7. A theme that is similar to that of figure 6 can also be observed in figure 7. First, tracer particles that are farther away from the solid boundary provides much better measurement accuracies than particles that are in the immediate vicinity of the solid boundary. Second, for small  $\Delta T$ , measurements based on near-wall particles significantly underestimate the actual external force, and this bias decreases as tracer particles become farther away from the solid boundary. Lastly, at large  $\Delta T$ , it is again observed that near-wall particle motions can lead to over-estimation of the external force, and this measurement bias is alleviated when force measurements



Figure 6. Force measurement accuracy derived from tracer particles in near-wall regions of various depths. Particles considered in the plot undergo hindered Brownian motions and experience an external force of  $F_x = 1$ .



Figure 7. Force measurement accuracy derived from tracer particles at various distances from the solid wall. Particles considered in this plot experience an external force of  $F_x = 1$ .

are conducted using particles that farther away from the solid boundary.

Together, figures 6 and 7 suggest that hindered Brownian motion can lead to force measurement inaccuracy, as evidenced by the fact that measurements taken on particles farther away from the solid surface are more accurate than the particles that are close to the wall. Furthermore, hindered Brownian motion can also lead to significant bias sampling and thus force measurement inaccuracy. It has been suggested that near-wall tracer particles tend to bias toward translation trajectories closer to the wall than farther away from the wall for small time duration between image acquisitions [12, 13]. Therefore for small  $\Delta T$ , near-wall particles undergoing hindered Brownian motion will tend to experience a larger of hydrodynamic drag. Consequently, the applied force will translate these particles less than one may anticipate based on the  $\beta_r$  values that one calculates using the initial Z positions. Lower overall ensembleaveraged particle translations then leads to under-estimation of the applied force magnitude. The opposite effect takes place at large  $\Delta T$ . Huang *et al.* [12] and Sadr *et al.* [13] had suggested that particles will tend to favor trajectories moving away from

the solid boundary for large  $\Delta T$  as they are now given enough time to venture out of their designated Z position range and still return during the period of  $\Delta T$ . As a result, they experience less overall hydrodynamic drag and translate more than they are anticipated to, leading to over-estimation of the applied force magnitude. These unique sampling biases are the results of near-wall tracer particles undergoing hindered Brownian unanticipated motion and experiencing changes in hydrodynamic drags, and therefore are absent in force measurements conducted using isolated tracer particles undergoing unhindered Brownian motion.

Still, our numerical investigations into near-wall particle tracking velocimetry reassure the notion that the relative dominance between the applied force and Brownian motion significantly impacts force measurement accuracies, as evidenced by figure 8. Larger applied force can be measured more accurately than small applied force can be for all  $\Delta T$ . However, the fact that Brownian motion is hindered for near-wall tracer particle can lead to more accurate force measurement at small  $\Delta T$  than measurements conducted with particles of unhindered Brownian motion presented in section 4.2.



Figure 8. Force measurement accuracy derived from tracer particles in the near-wall region of 1 < Z < 6.

## 5. CONCLUSION

In summary, using Langevin simulations we investigated the effects of unhindered and hindered Brownian motions on external force measurement accuracies derived from particle tracking velocimetry. It is found that the relative strength between the random forces that cause Brownian motion and the applied external force plays a critical role in measurement accuracy. It is expressed through both  $F_x$ , a constant characterizing the applied force and available thermal energy in the system, and  $\Delta T$ , the elapsed time between successive image acquisitions. We also found that hindered Brownian motion and the associated sampling trajectory biases contribute additional force measurement inaccuracies when particle tracking velocimetry is conducted in the vicinity of a solid boundary. Investigations in measurement bias reduction, bias-correcting algorithms and experimental verifications are currently underway.

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