Quantification of Brownian motion under presence of flow using Particle Diffusometry

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Abstract

Particle diffusometry (PD), a quantification method for the Brownian motion, is performed by recording temporally sequential images and using correlation analysis to obtain an ensemble diffusion coefficient for all particles captured in the imaging region (Clayton et al., 2017). PD is proven to be successful in the detection of the waterborne pathogen *V. cholerae* in environmental samples using different imaging techniques, including an inverted fluorescence microscope as well as a handheld hardware device operated with a smartphone (Clayton et al., 2019; Moehling et al., 2020). Although we intend to use PD to calculate diffusion coefficients in quiescent fluid, oftentimes unintentional fluid flows occur, creating measurement error when calculating the diffusion coefficient. In previous work, recordings under the presence of flow were discarded to avoid incorrect measurements of the sample. The diffusion coefficient is calculated as:

$$D = \frac{S_c^2 - S_a^2}{16M^2\Delta t} \tag{1}$$

Experimentally recorded image frames are obtained at an inter-frame time Δt . These frames are then parsed into interrogation windows. Interrogation windows from an image taken at Δt is correlated with itself to obtain autocorrelation peak width S_a . Two separate image frames recorded at t and t+ Δt are correlated to obtain cross-correlation peak width S_c . The S_c broadens compared to the S_a due to the presence of Brownian motion. The obtained peak widths can be then averaged spatially or temporally over number of image frames, to lower the random uncertainty of the measurement. The diffusion coefficient is then calculated using Eq. 1 (Olsen and Adrian, 2000), where M is the total magnification of the recording setup. In this body of work, we modify our approach in measuring the diffusion coefficient in the presence of fluid flow.

In the absence of flow, the correlation peaks described above produce axisymmetric Gaussian profiles. However, in the presence of flow, the correlation peak center shifts to the direction of the flow—exactly the principle behind PIV. If the flow also has gradients in it, the correlation peak stretches and rotates in the direction of shear, no longer axisymmetric. Since the Brownian motion along each axis is independent of others, proper identification of streamwise and cross-streamwise directions allow a generalized rotated two-dimensional elliptical Gaussian function to be fit to the cross-correlation (Chamarthy et al., 2009). The diffusion coefficient can be calculated from the cross-streamwise correlation peak width.

In this work, simulated Brownian motion images were generated with a frame rate of 15 frames per second with a diffusion coefficient of $1 \times 10^{-12} m^2/s$ with a variety of conditions: 0 to 80 degrees rotation with 10-degree increments, 1 to 10 pixel/ Δt flow velocity with 10 pixel/ Δt increments, and three different flow types (uniform, Couette, and Poiseuille flows). Once the correlation peaks were calculated, all the peaks from each of the interrogation windows were averaged temporally over 100 frames image sets to reduce statistical random errors. A 3-point Gaussian peak fit was applied in both x- and y-directions to determine the sub-pixel maximum location for further flow analysis. To calculate the cross-streamwise peak width, a total of 11×11 pixel² areas were fitted (in contrast to the typical 5-point PIV peak fit), and the ellipse equations at e^{-1} height is calculated using the fitted Gaussian profile. Resulting diffusion coefficient values of varying angles and velocities were averaged with respect to the induced velocity values.

As shown in Fig. 1(a), a 3-point Gaussian sub-pixel fit on a single pass correlation poses no difficulty in estimating the flow velocity from the generated simulation videos. A larger measurement variation occurred

in the low flow velocity region (i.e. 1-4 pixel/s), where distinguishing between the flow velocity and the Brownian motion of particles presents a greater margin of error. For the case of measuring particle diffusion coefficient, presented in Fig. 1(b), the diffusion coefficient calculated in the presence of uniform flow show minimal deviation from their simulated values, hence the smaller error. However, with non-uniform flows, we see greater deviations from theoretical values due to the gradient of the flow. Overall, an increase in error is observed as the flow velocity is increased. Analysis reveals that with the developed algorithm, analyzed diffusion coefficients show an error greater than 10% when the flow velocity exceeds 8 pixel/ Δt .

Experimentally, flows were generated using a syringe pump through a rectangular channel. As the flow profile forms an elongated Poiseuille flow along the wider dimension of the channel, the focal point is adjusted to ensure the recorded flow profile is that of a uniform flow. Simulation parameter for the uniform flow is extended to 22 pixel/ Δt to better compare the trend between the simulation and experiment results. Due to the difficulty of obtaining even increment of volumetric flow, x-directional error bar is incorporated to account for the variation. The resulting Fig. 1(c) shows the experimental data following the trend of the simulation results. The experiment shows the analyzed diffusion coefficients results in an error greater than 10% when the flow exceeds 18 pixel Δt with the developed algorithm. These 18 pixels account for 17% of overall interrogation window. In other words, the velocity is equivalent to $6.6\mu m/\Delta t$, a significant movement if we take into account the particle size of 470*nm* that is used for the experimental verification. This current body of work shows a promising initial work toward measuring diffusion coefficients in the presence of fluid flow.



Figure 1: Comparison of simulation results with analyzed values (a,b), and with experiment values (c).

References

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