

## Supporting Information

### Imaging Fluorescence Correlation Spectroscopy Studies of Dye Diffusion in Self-Assembled Organic Nanotubes

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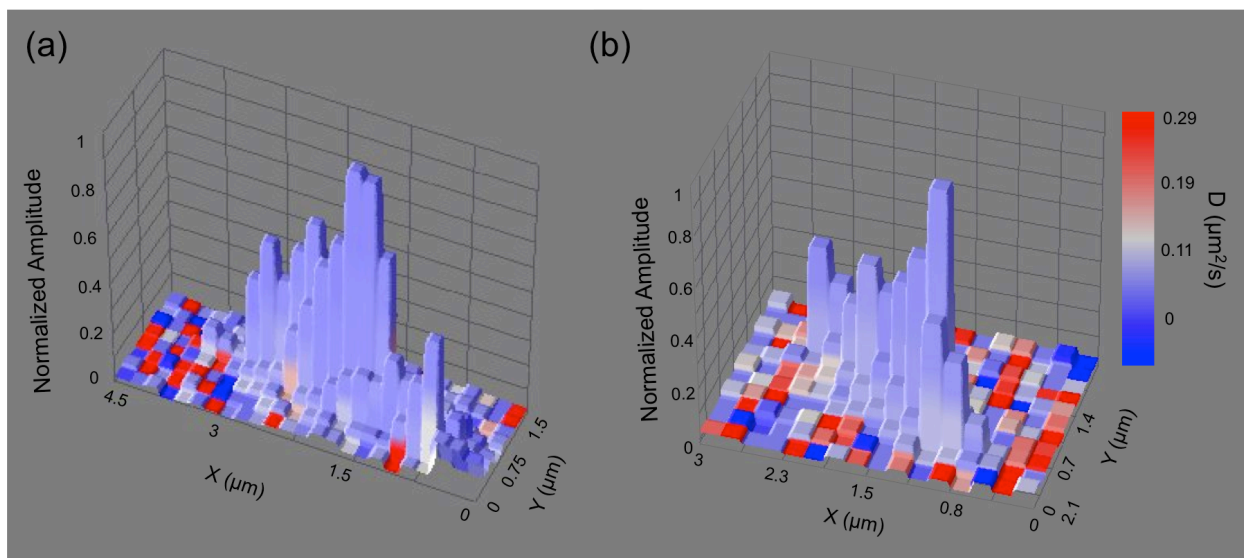
Additional details on the temporal resolution of the imaging-FCS method are provided. Also included are two additional 3D surface plots of two nanotubes showing autocorrelation amplitude and diffusion coefficient information. A detailed discussion of the spatial dependence of dye diffusion dynamics is given. A representative imaging-FCS video of one nanotube is also provided.

## Temporal Resolution

Several experimental factors limit the range of diffusion coefficient values that can be measured in this work. As noted in the experimental section, the videos were acquired with  $\sim 6$  ms time resolution (exposure + read time). The lateral dimension of the detection volume was estimated from a convolution of the pixel size ( $0.1875 \mu\text{m}$ ) with the Gaussian point spread function width ( $0.240 \mu\text{m}$ ) of the microscope.<sup>1</sup> In this case, the largest diffusion coefficients that can be measured by this method are  $\sim 5 \mu\text{m}^2/\text{s}$ . This is significantly smaller than the  $\sim 470 \mu\text{m}^2/\text{s}$  expected for SRB diffusion in bulk solution.<sup>2</sup> Therefore, only the relatively slow diffusion of SRB molecules interacting with the nanotubes can be probed in these studies.

## 3D Surface Plot Models

Figure S1 depicts two additional 3D surface plots showing composite images of the autocorrelation amplitude and diffusion coefficient data. Similar to Figure 4, the amplitudes are approximately zero off the nanotube and the diffusion coefficients obtained are dominated by noise in these regions. Meaningful diffusion coefficient data are only obtained from the nanotubes themselves, for which the uniform color depicts only relatively small variations in these values. Along with the data shown in Figure 4, these three composite images of the nanotubes reveal the consistency of the diffusion coefficient data obtained from different tubes. These same data also demonstrate that there is no clear position dependence in the concentration of diffusing SRB molecules in the nanotubes.

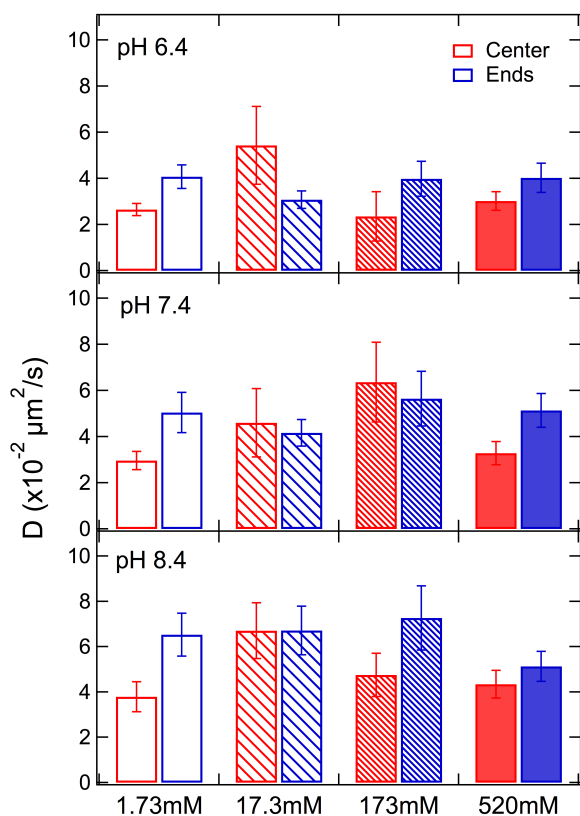


**Figure S1:** 3D surface plots of two additional nanotubes, showing the normalized autocorrelation amplitude as height, with the measured diffusion coefficient depicted by the color scale.

### Spatial Dependence of SRB Diffusion Dynamics

The position dependence of SRB diffusion along the nanotubes was characterized by averaging the SRB diffusion coefficients for the nanotube centers and ends. Each nanotube was equally divided into three regions (two ends and the center) along the long axis of the tube. The mean diffusion coefficient for each region was then obtained by averaging the  $D$  values of each pixel in one region. Figure S2 plots the diffusion coefficients obtained from center and ends of the nanotubes under different pH and ionic strength conditions. No clear trends are observed in these data. Statistically, only five cases out of twelve show differences in  $D$  at  $> 80\%$  confidence, while opposite trends are frequently observed between the tube ends and center. It is concluded that the diffusion dynamics are relatively homogeneous along the full length of the tubes. This result contrasts with those obtained from lipid nanotubes in which the rate of diffusion was found to be faster at the tube ends and slower in the nanotube center.<sup>3</sup> In the

latter, the observed differences were attributed to heterogeneous packing of lipids. The results obtained from our present nanotubes are consistent with a homogeneous nanotube structure and homogeneous filling of the nanotubes with aqueous solution (at least on resolvable length scales).



**Figure S2:** Averaged apparent diffusion coefficients obtained from tube center and ends under varied pH and ionic strength conditions. Error bars represent the standard deviation on the mean.

**Video S1.** Fluorescence video (20000 frames, 167 frames/s) depicting imaging-FCS measurement on one nanotube. Clear photobleaching in the long time range can be observed in the video. The data shown in Figure 2 were derived from this video.

## REFERENCES

- (1) D. Boening, T. W. Groemer, J. Klingauf, Applicability of an EM-CCD for Spatially Resolved TIR-ICS, *Opt. Express*, 2010, **18**, 13516-13528.
- (2) D. A. Sabatini, Sorption and Intraparticle Diffusion of Fluorescent Dyes with Consolidated Aquifer Media, *Ground Water*, 2000, **38**, 651-656.
- (3) L. Guo, P. Chowdhury, J. Fang, F. Gai, Heterogeneous and Anomalous Diffusion inside Lipid Tubules, *J. Phys. Chem. B*, 2007, **111**, 14244-14249.