Supplementary Information

Spectroscopic Imaging Studies of Nanoscale Polarity and Mass Transport Phenomena in Self-Assembled Organic Nanotubes

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Additional details on the measurements of the NR-OH pKₐ are provided, along with representative nanotube video data.

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Determination of $pK_a$ of NR-OH dye

A pH titration experiment was conducted to determine the $pK_a$ of the phenolic proton on the NR-OH dye. The $pK_a$ of the protonated amine groups on the nanotube inner surface was previously reported to be 7.27 in aqueous solution.\textsuperscript{1} To directly compare with this $pK_a$, the present titration was performed in water as well. During the titration, small aliquots of 0.2 wt% NaOH aqueous solution were added to the NR-OH dye solution continuously. The pH was measured using a pH meter after each addition of NaOH. The NR-OH absorption spectra were recorded on a UV-vis spectrometer during the titration.

As a solvatochromic dye, the absorption peak of NR-OH shifts as the polarity of the environment changes. It has been reported earlier that the absorption peak of NR-OH shifted upon the addition of 9.9 mM tetrabutylammonium hydroxide (Bu\textsubscript{4}NOH) in methanol.\textsuperscript{2} The peak shift was assigned to the deprotonation of the phenolic OH group on NR-OH in the presence of base. Thus, in this experiment, we determined the $pK_a$ of NR-OH by plotting the peak absorption wavelength as a function of pH. Figure S1 shows the absorption peak shift upon the addition of base. The $pK_a$ of the phenolic proton was determined to be 9.21 from these data. The relatively large $pK_a$ of the phenolic proton on NR-OH suggests that it will not be deprotonated by interaction with the amine groups on the tube inner surface.

![Figure S1](image_url)

\textbf{Figure S1} The NR-OH absorption peak shift as a function of solution pH.
**Video S1.** Fluorescence video (20000 frames, 149 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 6 were derived from this video.

**REFERENCES**
