Supporting Information

Modified Organosilica Core-Shell Nanoparticles for Stable pH Sensing in Biological Solutions

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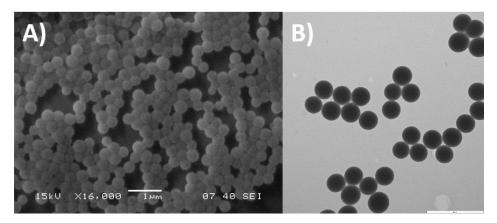
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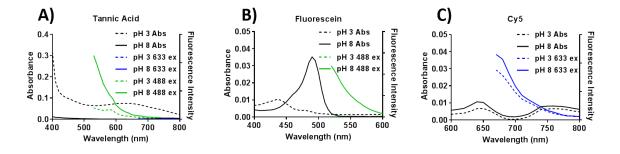
Continuous monitoring, organosilica, nanosensor, tannic acid, pH measurement

Before the second step involving shell addition and using the previously published protocol the standard deviation in particle size was found to be less than 10% consistent with results presented by Miller et al. (Supp. Figure 1)



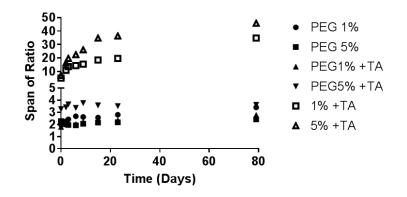
Supp. Figure 1:Electron Microscopy Unmodified Distribution. A) SEM of MPS-Cy5 30 minute incubation B) TEM of MPS-Cy5 particles 30 minute incubation (scale bar 1000 nm)

The absorbance and background fluorescence of the porogen TA was interrogated and it was found that there were broad absorbance bands overlapping the excitation and emission wavelength of both the chosen fluorophores (Supp. Figure 2). This overlap helps to explain the effect of TA on the fluorescence of the fluorophores and suggests more complicated mechanisms of interaction between the molecules. The pH dependence of fluorescein and independence of Cy5 was also observed.



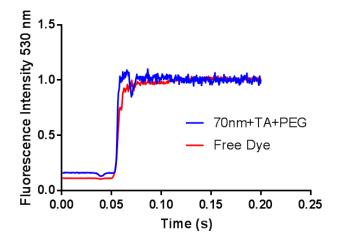
Supp. Figure 2: Absorbance of TA and Emission of Fluorescein and Cy5. A) Absorbance (Abs) and emission using excitation wavelengths of 488 and 633 nm (488 ex and 633 ex) at both pH 3 and 8 of TA B) Absorbance and emission using an excitation wavelength of 488 nm at both pH 3 and 8 of fluorescein C) Absorbance and emission using an excitation wavelength of 633 nm at both pH 3 and 8 of Cy5

To show the stability of paricles the span of the sigmoidal interpolations was graphed over time (Supp. Figure 3). The span of the calibration curves produced using sensors containing TA plateau over time suggesting that the sensors reach an equalibrium. Sensors with PEG conjugated display span stability even when incubated with TA.



Supp. Figure 3: Span of Calibration Curves over Time. The spans of the sigmodial interpolations of the pH sensitivity data shown in Figure 5 overtime.

Using a SX20 Stopped Flow Spectrometer in a single mixing configuration the measurement of the response time of the produced sensors was attempted however the kinetics of the mixing, pH response of free dye and pH response of nanosensors was virtually indistinguishable (Supp. Figure 4) suggesting that the difference between them could not be resolved using this instrument. Experiments consisted of combination of sample suspended in 0.01 M citric acid with a 0.2 M Sodium Phosphate solution pH 8. FITC filters were used and measurements were pre-triggered and taken over 1 second.



Supp. Figure 4: Response Time of Free Dye vs Sensors. This figure shows the change in fluorescent signal as either particles or free dye initially in acidic conditions are mixed with a basic buffer.