

## Supporting Information

### Soft Microcapsules with Highly Plastic Shells Formed by Interfacial Polyelectrolyte-Nanoparticle Complexation

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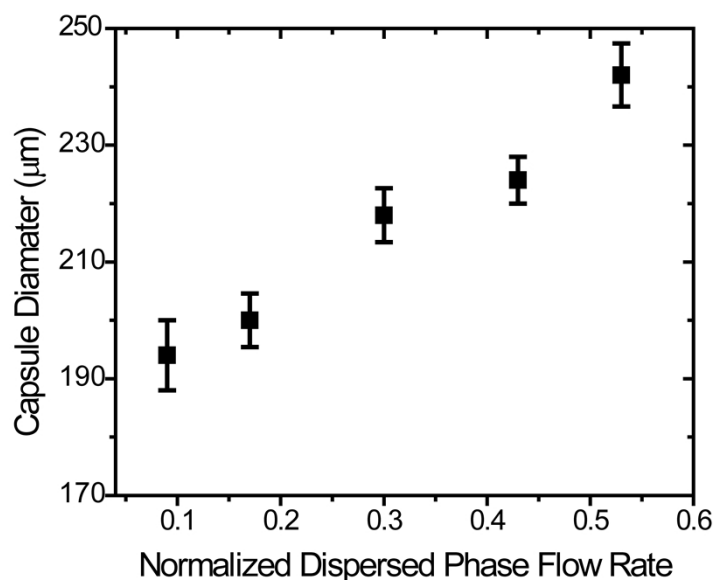
#### **Materials**

Polystyrene-block-poly(ethylene-ran-butylene)-block-polystyrene (s-SEBS, 29 wt% styrene, 55-56% sulfonated), alumina coated silica nanoparticles (LUDOX-CL), and lysozyme were obtained from Sigma Aldrich and used as nanoparticles or proteins, and polyelectrolyte in the aqueous and organic phase, respectively. The molecular weight of the s-SEBS was not provided by the manufacturer. The zeta potential for 0.1 wt%, pH 5.8 silica nanoparticles and 0.1 wt% lysozyme pH 5.5 nanoparticles under conditions relevant to our study are  $\sim 60$  mV<sup>1</sup> and  $\sim 8$  mV<sup>2</sup>, respectively.

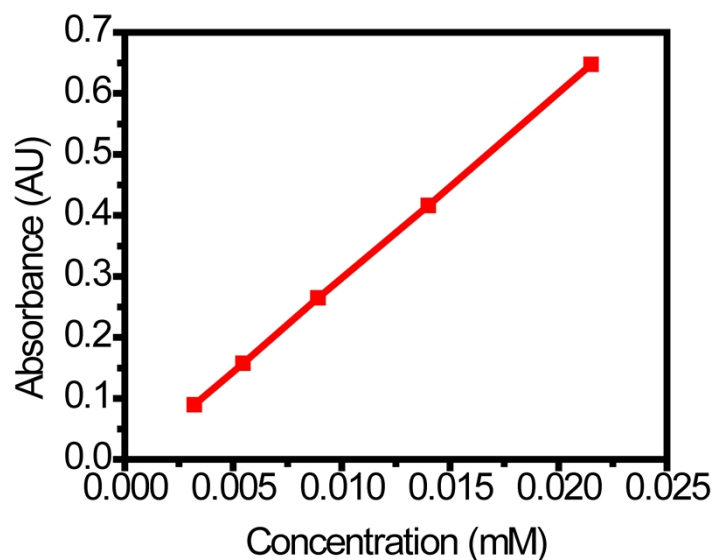
Toluene (Sigma Aldrich) served as organic solvent for s-SEBS. Tetraethyl orthosilicate (TEOS) and fluorescein isothiocyanate (FITC), ammonium hydroxide, and ethanol were obtained from Sigma Aldrich and used to fabricate amine modified silica nanoparticles. Oil Blue N was obtained from Sigma Aldrich and used as the dye for release studies.

#### **UV-Vis Measurements**

Monodisperse microcapsules (243  $\pm$  4.6  $\mu$ m diameter) containing the dye were collected in a vial for 12 min. The vial was tightly sealed and aliquots of the supernatant are taken at set times and spectra measurements are recorded using Varian UV-VIS. Spectra measurements are converted to dye concentration based on a calibration curve



**Figure S1.** (a) The effect of dispersed fluid rate flow rate on capsule diameter. The dispersed phase flow rate was normalized by the continuous phase flow rate.



**Figure S2.** Calibration curve for Oil Blue N dye in toluene. Absolute absorbance recorded by UV-Vis as a function of dye concentration in toluene.

## References

1. Boussu, K.; Belpaire, A.; Volodin, A.; Van Haesendonck, C.; Van der Meeren, P.; Vandecasteele, C.; Van der Bruggen, B., Influence of membrane and colloid characteristics on fouling of nanofiltration membranes. *Journal of Membrane Science* **2007**, 289, (1–2), 220-230.
2. Bumiller, M. New ISO Standards for Zeta Potential Analysis [http://www.labmate-online.com/article\\_read/1170/](http://www.labmate-online.com/article_read/1170/)

