

# Micro- and nanoscale hierarchical structure of core-shell protein microgels

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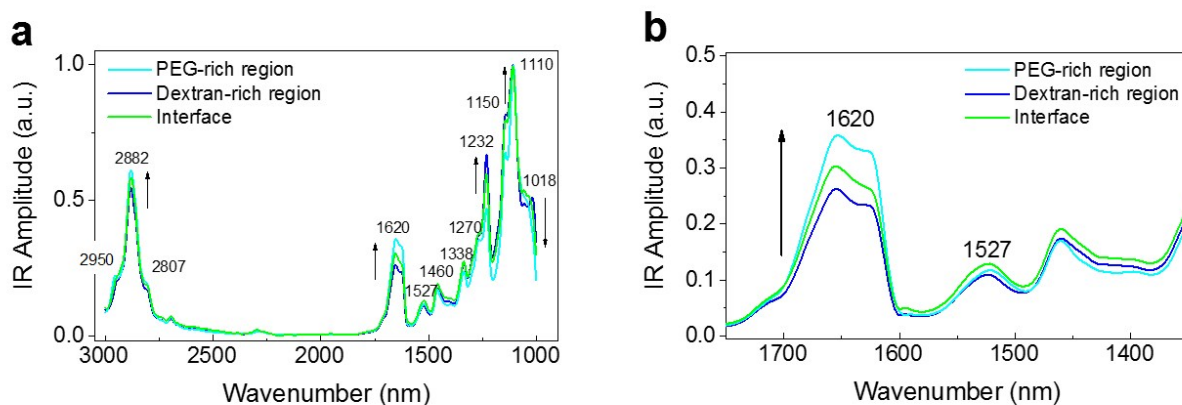
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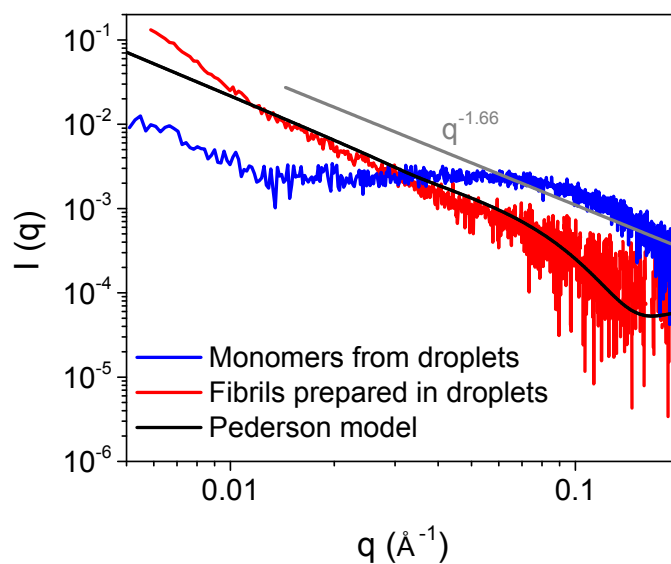
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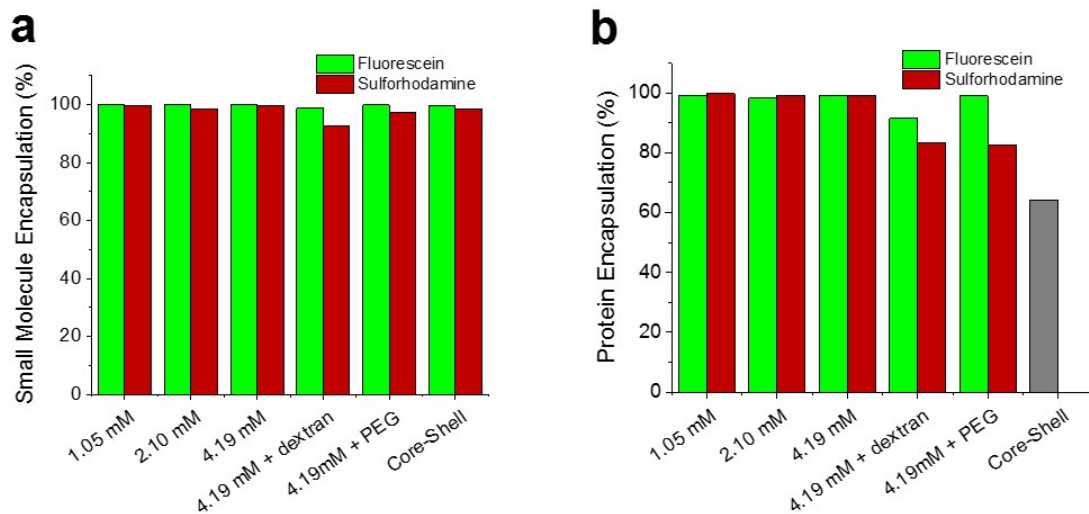
SUPPLEMENTARY INFORMATION



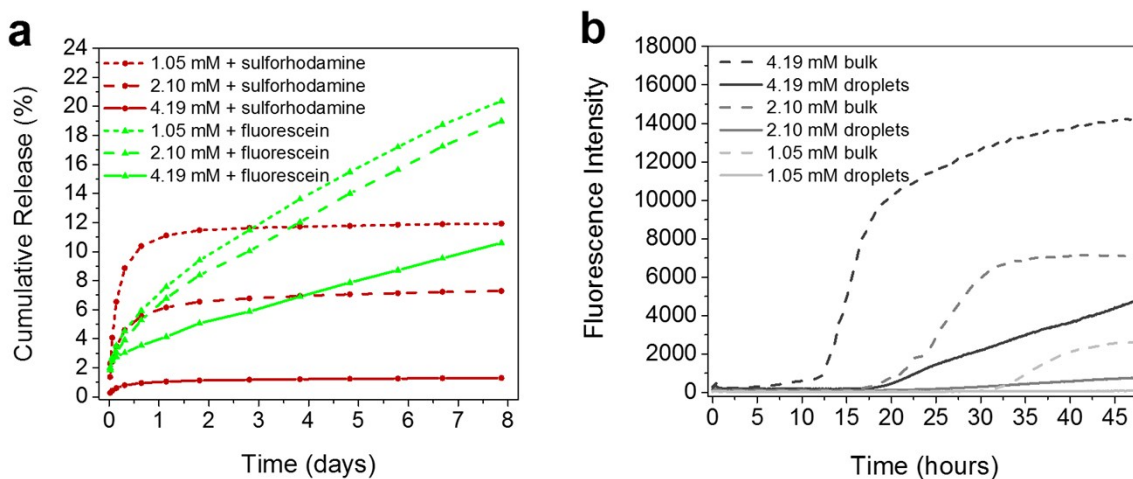
**Figure S.1.** AFM-NanoIR infrared spectroscopy of PEG-rich phase, dextran-rich phase, and contact region of core-shell microgels. (a) IR spectrum and (b) key peaks of spectrum, indicating an increase in  $\beta$ -sheet content with increasing PEG concentration.



**Figure S.2.** Small angle X-ray scattering fit with Pederson model. Monomers and fibrils extracted from incubated and non-aggregated droplets, respectively, showed results similar to those obtained from monomers and fibrils in microdroplets with the absence of  $q^{-4}$  slope at low  $q$ .



**Figure S.3.** Encapsulation efficiency of (a) small molecules and (b) lysozyme in single-shell and core-shell microgels.



**Figure S.4.** Effects of lysozyme concentration on release kinetics and fibrillar content of microgels. (a) Release profiles of small molecules in single shell microgels. (b) Thioflavin T fluorescent emission profiles of bulk protein and protein microgels.

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