Supporting Information

Materials. Peptide synthesis was by F-moc chemistry, and product analysis by high-performance liquid chromatography and mass spectrometry.

Microcapsule preparation. Designed peptides were dissolved separately in 10 mM Tris-HCl buffer solution, pH 7.4 to a final concentration of 1 mg/mL. Melamine fluoride (latex) particles were prepared for assembly by rinsing with water and introduced into the negatively-charged polypeptide solution first. The sample was mixed manually, kept at room temperature for 20 min with occasional agitation, and then centrifuged at 1143*g* for 1 min. Coated melamine latex particles were introduced into the positively-charged polypeptide solution in the same way. These steps were repeated until the desired number of layers was formed. The latex core was dissolved in NaCl-HCl buffer (100 mM, pH 1.6).

Fluorescent labeling. Poly-(L-lysine) labeled with fluorescein isothiocyanate (FITC) was used as the outermost layer of "locked" and "unlocked" capsules for visualization using fluorescence microscopy. FITC was dissolved in dimethylsulfoxide (DMSO), and poly-(L-lysine) in 0.1 M carbonate buffer at pH 9. The FITC solution was added to the poly-(L-lysine) solution in small aliquots with gentle shaking, and the resulting mixture was incubated overnight at 4 ºC. Excess FITC was removed by extensive dialysis against carbonate buffer. Capsules were rinsed thrice using deionized water and centrifugation to remove excess FITC-poly-(L-lysine).

Microcapsule characterization. Confocal laser scanning micrographs were obtained with a

Leica TCS NT inverted confocal system (Germany) equipped with a 100x oil-immersion objective of numerical aperture 1.4. FITC-labeled capsules were readily visualized by fluorescence microscopy using a Nikon inverted microscope Eclipse TS 100/TS100-F (Japan).