

## **Supporting Information**

### **Morphology control and surface functionalization of Protein-SiO<sub>2</sub> hybrid capsules**

Huihui Wang,<sup>a‡</sup> Tayebah Mirzaei Garakani,<sup>a‡</sup> Tim Krappitz,<sup>a</sup> Patrick van Rijn,<sup>\*a,b</sup> and Alexander Böker<sup>\*a</sup>

<sup>a</sup> *DWI an der RWTH Aachen e.V., Lehrstuhl für Makromolekulare Materialien und Oberflächen, RWTH Aachen University, Forckenbeckstrasse 50, D-52056 Aachen, Germany. Fax: +49-241-8023317; Phone: +49-241-8023304; E-mail: boker@dwi.rwth-aachen.de*

<sup>b</sup> *Department of Biomedical Engineering-FB40, W.J. Kolff Institute for Biomedical Engineering and Materials Science, University of Groningen, University Medical Center Groningen, A. Deusinglaan 1, 9713 AV Groningen, The Netherlands. Phone: +31-50-3633141; E-mail: p.van.rijn@umcg.nl*

**S1: Interfacial tensiometry**

**S2: SEM-analysis emulsion templated mineralization using only CTAB**

**S3: Normalized Absorption of calcein modification**

**S4: Supernatant analysis of modified structures with calcein**

### S1: Interfacial tensiometry

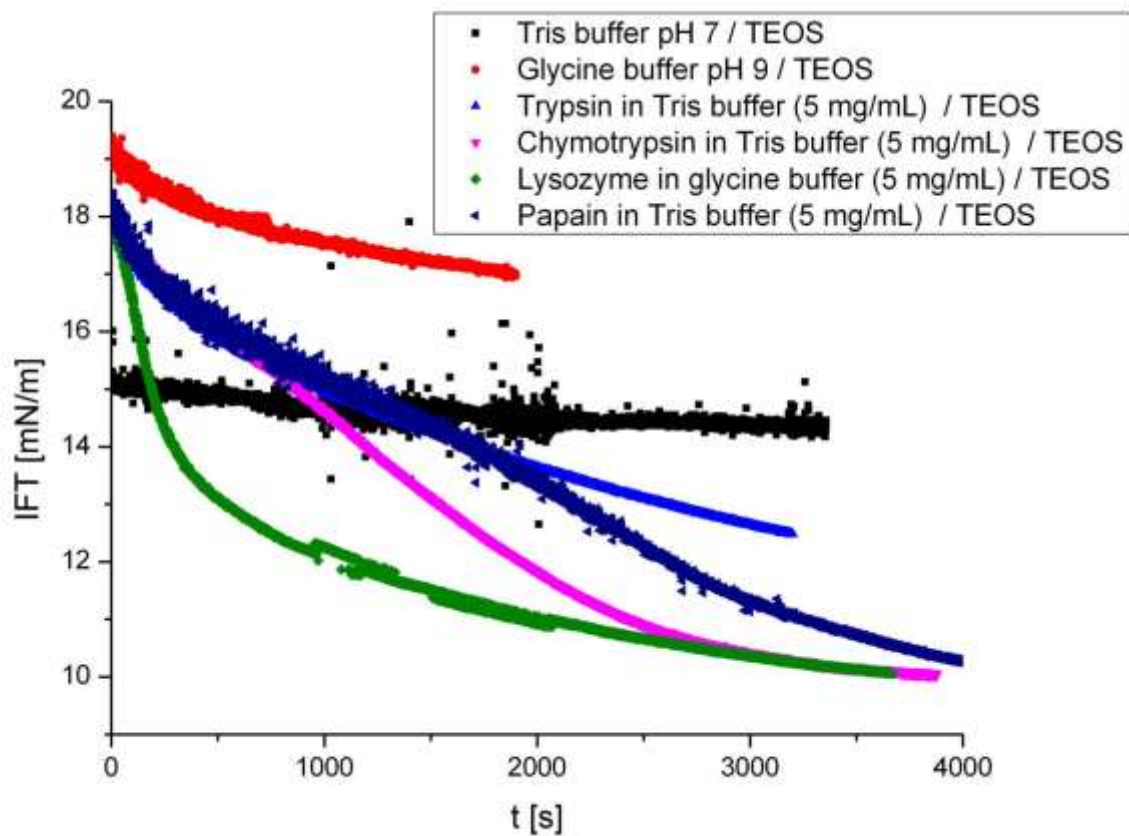


Figure S1: Interfacial tension measurements of interface between pure buffers and TEOS and between different protein solutions and TEOS.

## S2: SEM-analysis emulsion templated mineralization using only CTAB

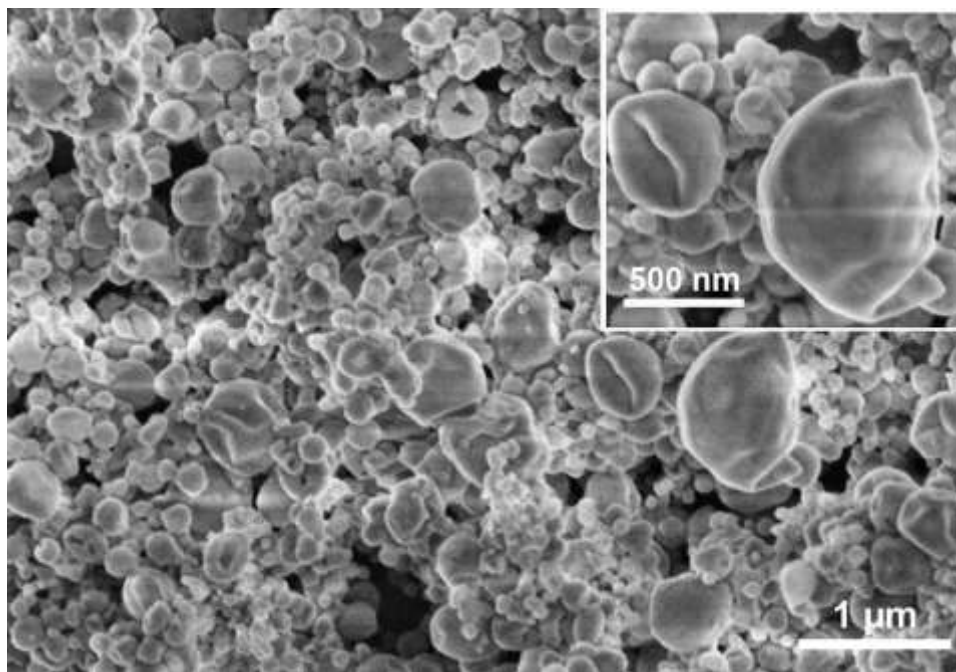


Figure S2: SEM images of SiO<sub>2</sub> capsules prepared via the TSA method using glycine buffer without lysozyme but only with CTAB. This provides insights in the contribution of CTAB with respect to mineralization reactivity and structure formation. Soft shells are found which are smooth and no specific fine-structure. This indicates that the specific combination of lysozyme and CTAB is needed for the folded structures shown in Figure 4 C/F of the main text.

### S3: Normalized Absorption of calcein modification

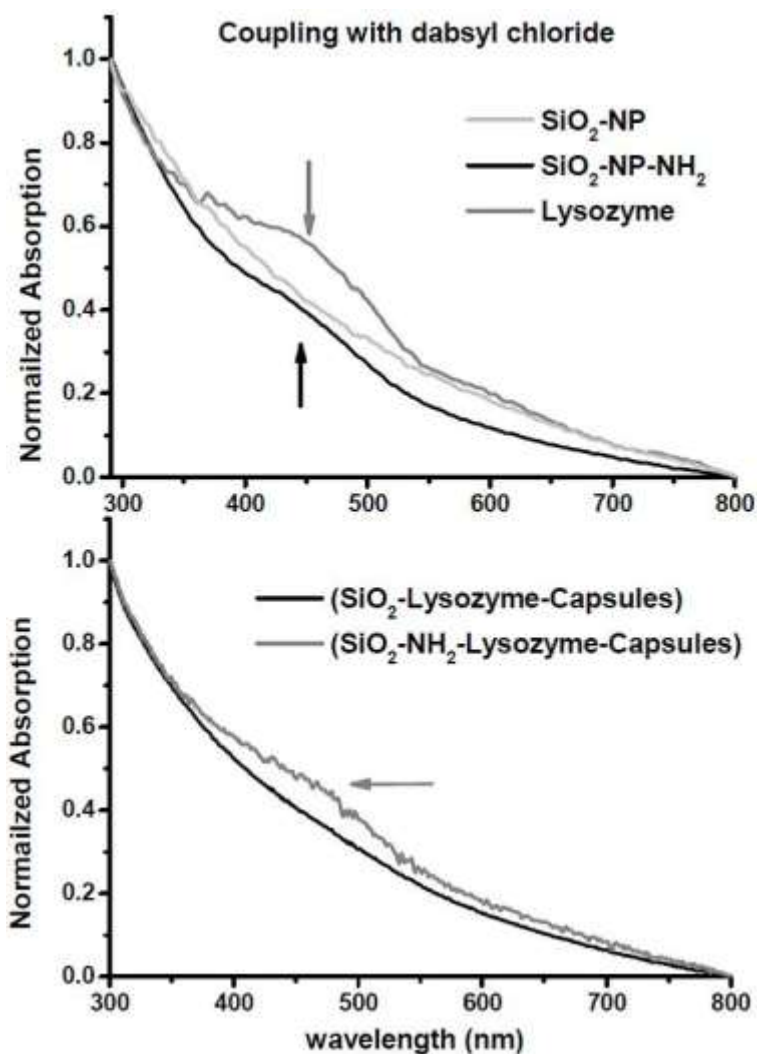


Figure S3: Absorption spectra of with dabsyl chloride modified structures: SiO<sub>2</sub>-nanoparticles; SiO<sub>2</sub>-NH<sub>2</sub> nanoparticles; Lysozyme (top); SiO<sub>2</sub>-lysozyme capsules prepared according to the CA method as described in the main text using only TEOS and SiO<sub>2</sub>-NH<sub>2</sub>-lysozyme capsules prepared via the same approach but with a TEOS/APTMS mixture of ratio 98/2 (bottom).

### S4: Supernatant analysis of modified structures with calcein

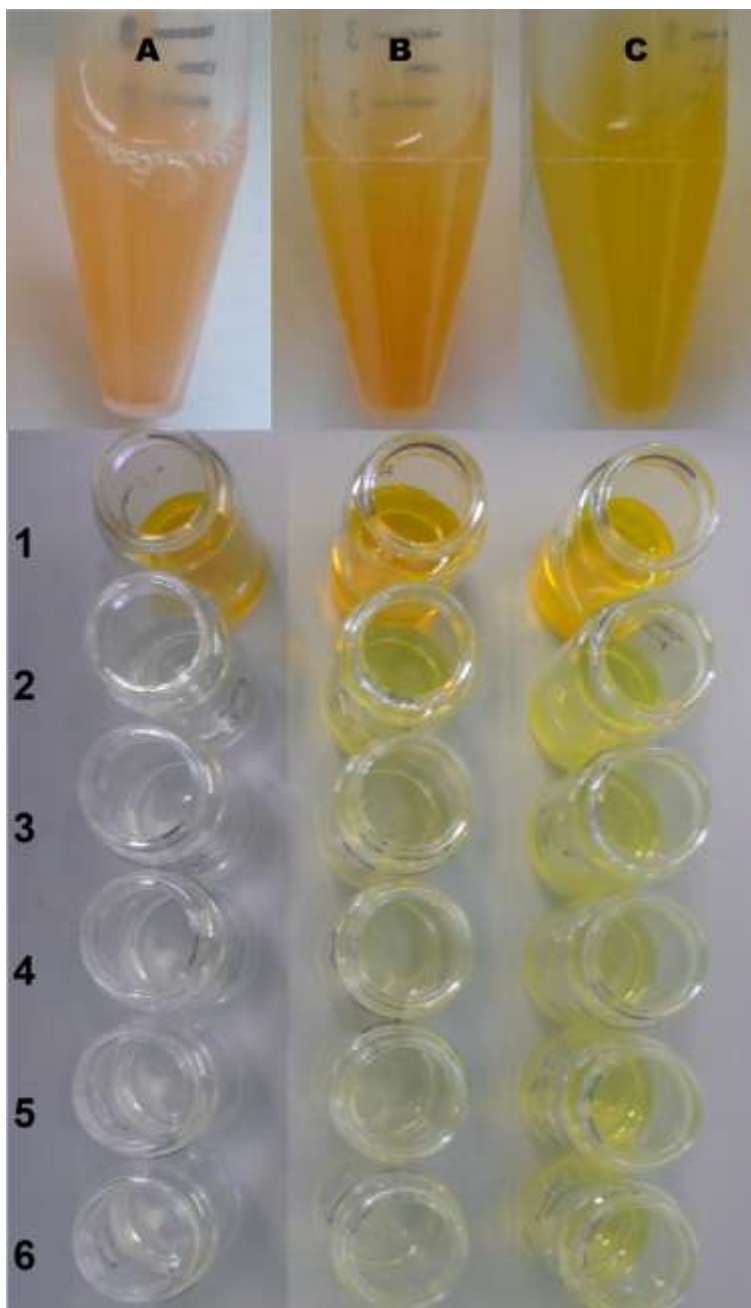


Figure S4: Supernatant collected after each washing step for : A) silica nanoparticle modified with APTMS; B) silica capsules modified with APTMS; C) silica capsules without modification.