Electronic Supplementary Material (ESI) for Soft Matter

Assembly of the anisotropic microcapsules in aqueous dispersions

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Table S1. Free energy components of liquid and solid surfaces used in calculations.

	Surface free energy (mJ/m^2)			
Material	γ_s	γ_s^{LW}	γ_s^+	γ_s^-
water[1]	72.8	21.8	25.5	25.5
PMAA[2,3]	41	40	0	9

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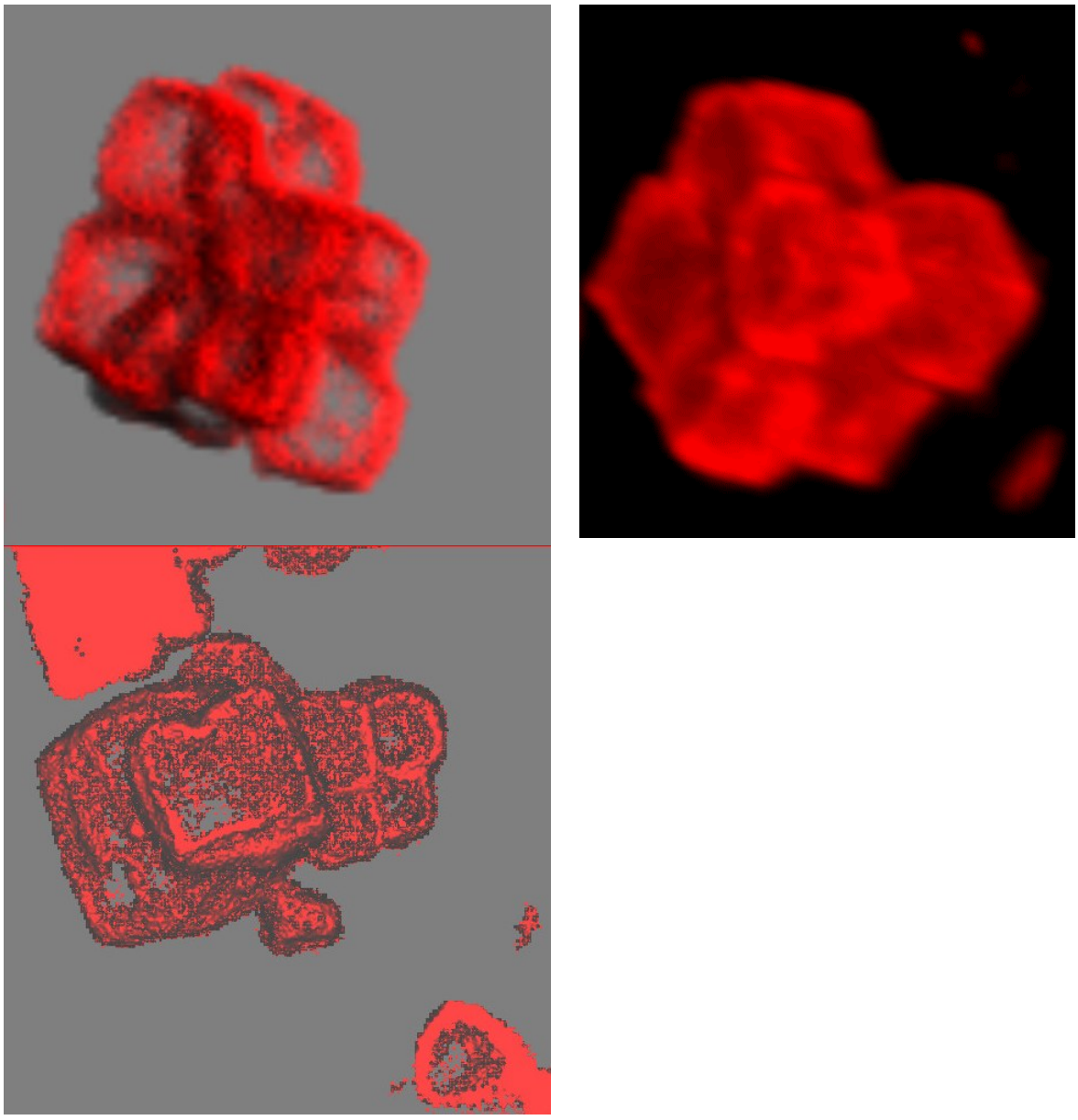

Figure S1. Confocal images of three dimensional (3D) assemblies of cubic $(PVPON/PMAA)_4$ microcapsules in buffer pH 4.5, C=0.1 M.

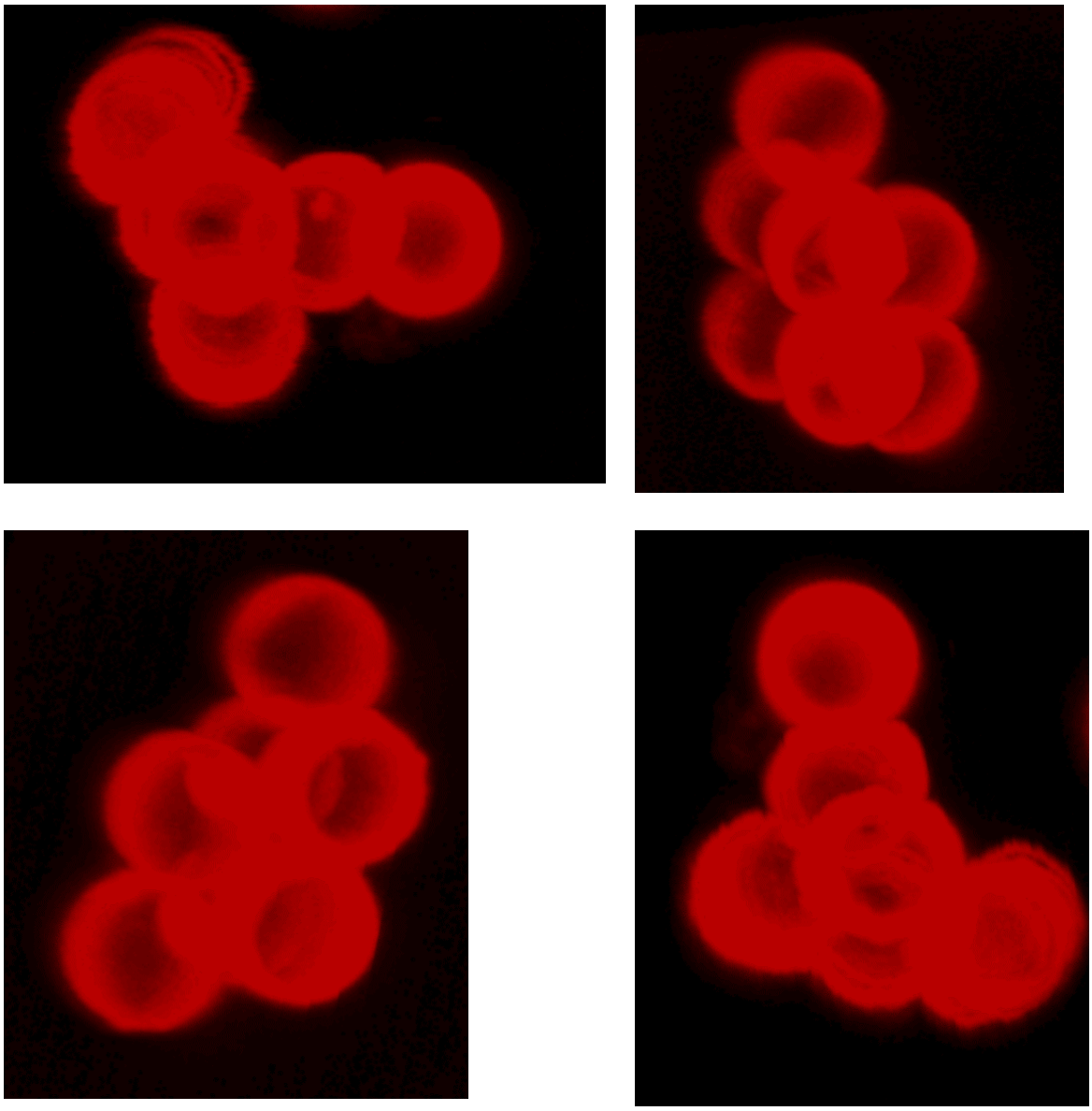

Figure S2. Confocal images of three dimensional (3D) assemblies of spherical $(PVPON/PMAA)_4$ microcapsules in buffer pH 4.5, C=0.1M NaCl

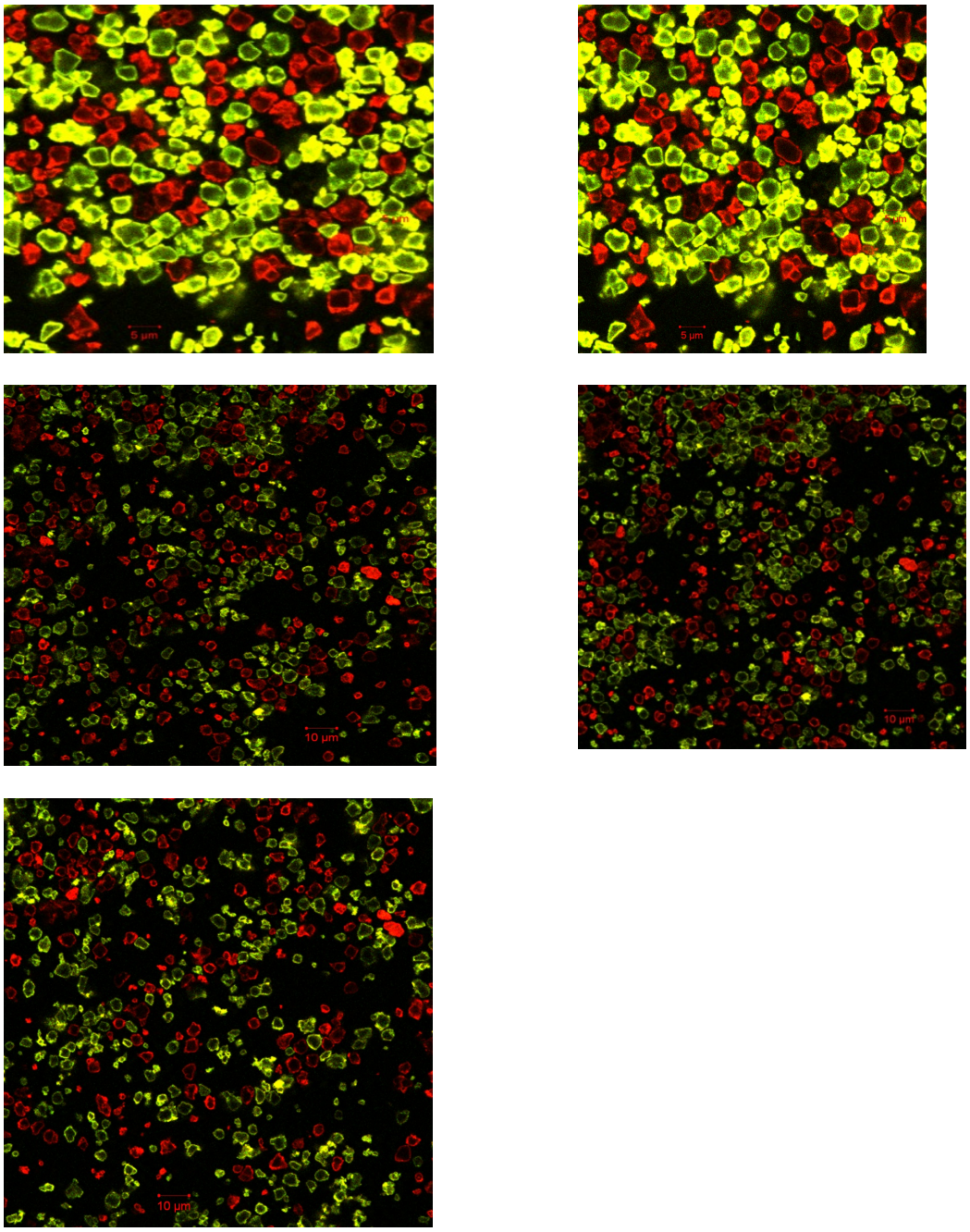

Figure S3. Confocal images of assembled cubic $(PVPON/PMAA)_4$ and $(PVPON/PMAA)_4PPE$ microcapsules in buffer pH 3.5, C= 0.01 M NaCl

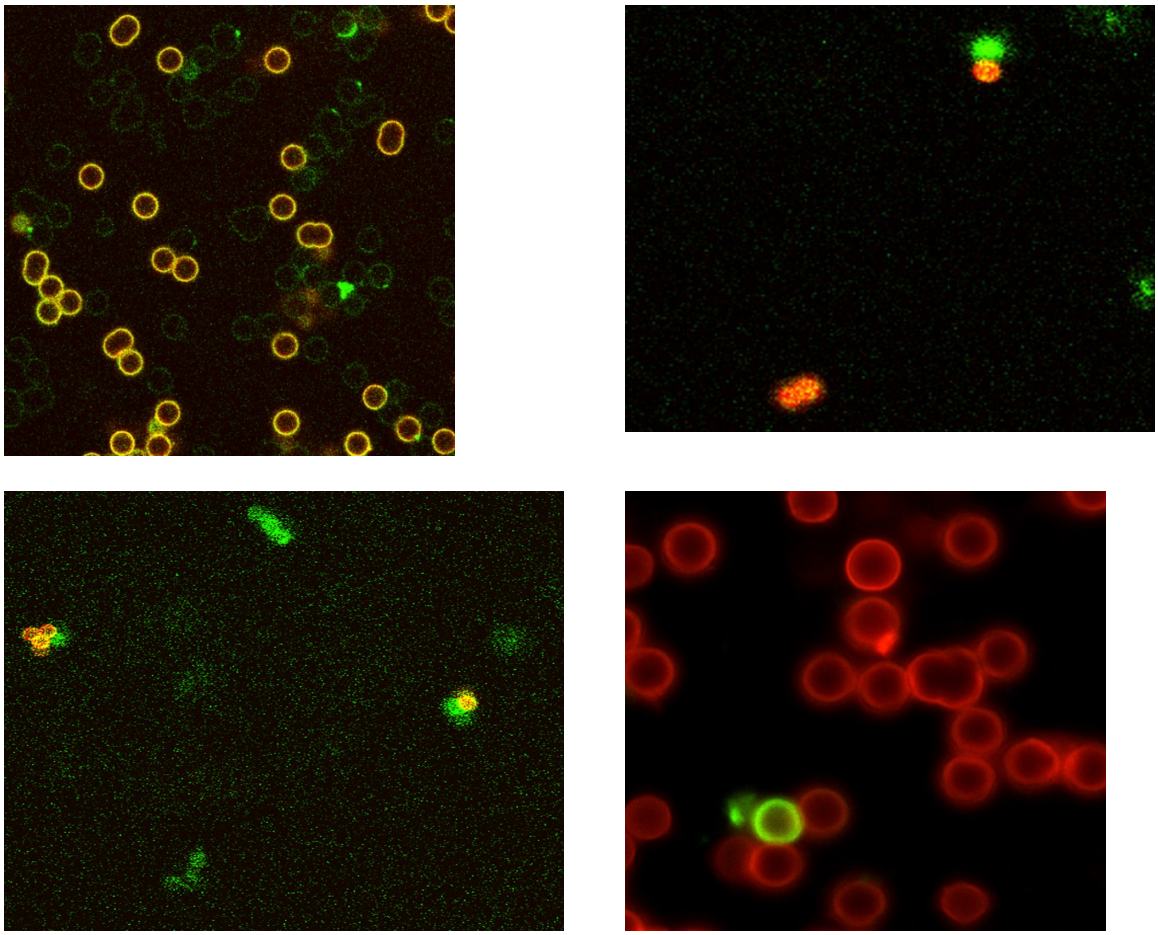

Figure S4. Confocal images of assembled spherical $(PVPON/PMAA)_4$ and $(PVPON/PMAA)_4PPE$ microcapsules in buffer pH 3.5, C= 0.01 M NaCl

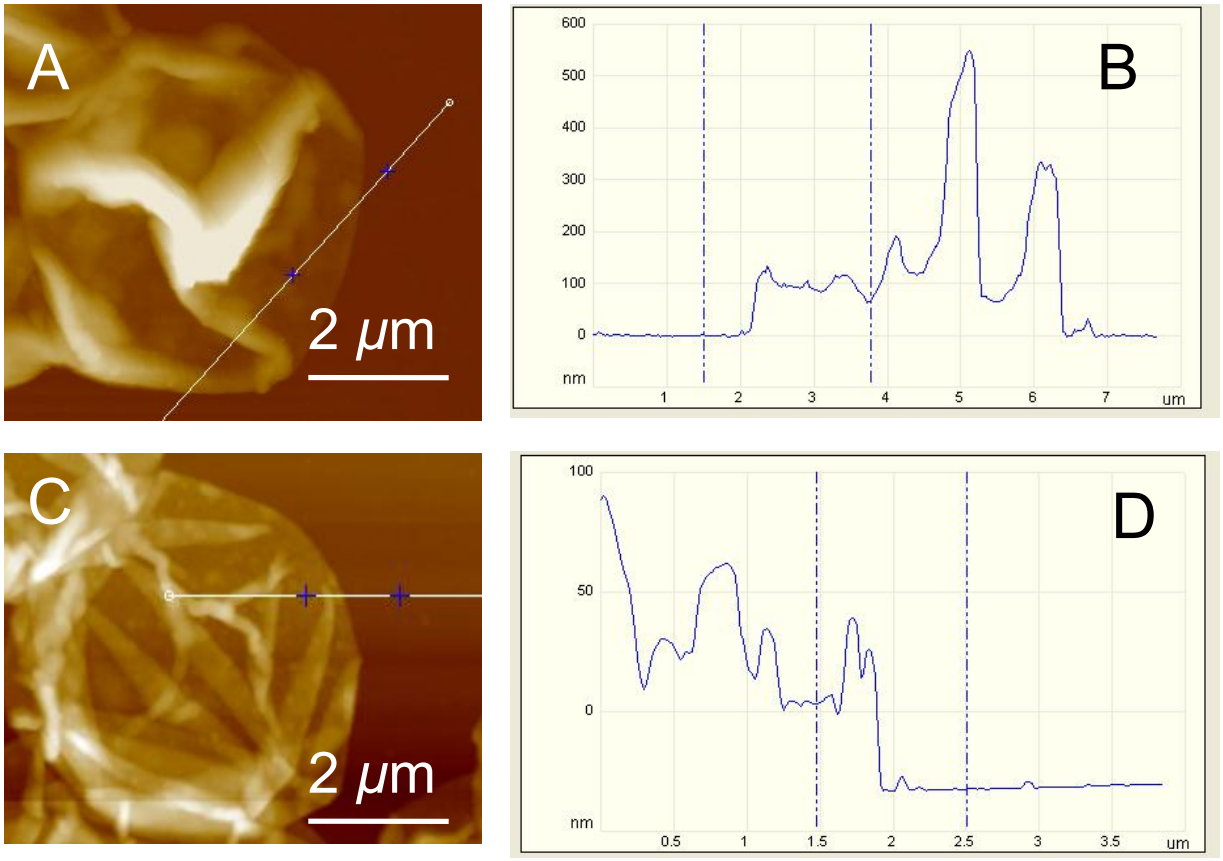

Figure S5. AFM cross section analysis of cubic (A, B) and spherical (C, D) $(PVPON/PMAA)_4$ microcapsules. Thickness of bilayer was estimated from flat areas in cross-sections.

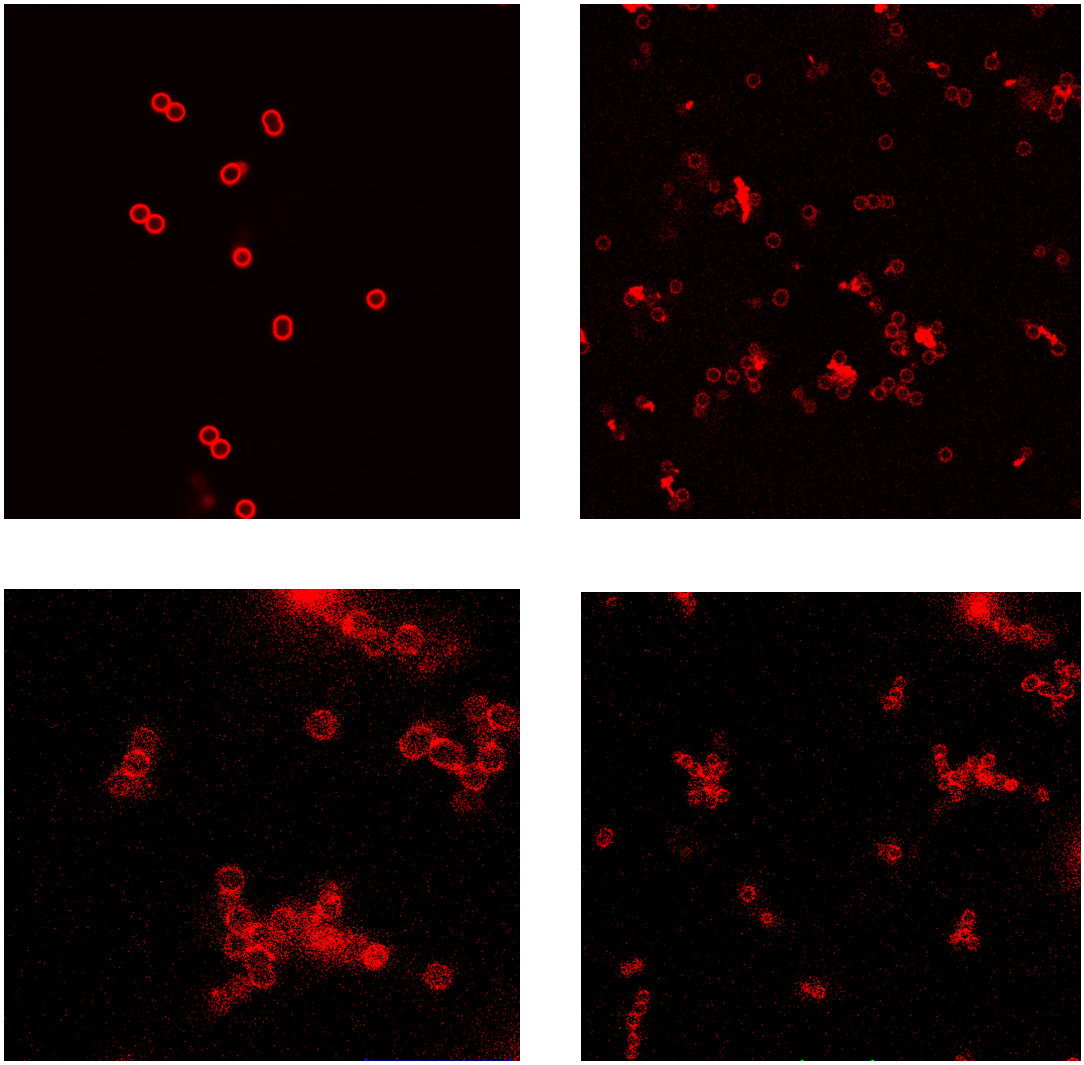

Figure S6. Confocal images of spherical $(PVPON/PMAA)_4$ microcapsules assemblies in buffer pH 4.5 C=0.01 M NaCl (top) and C=1 M NaCl (bottom).

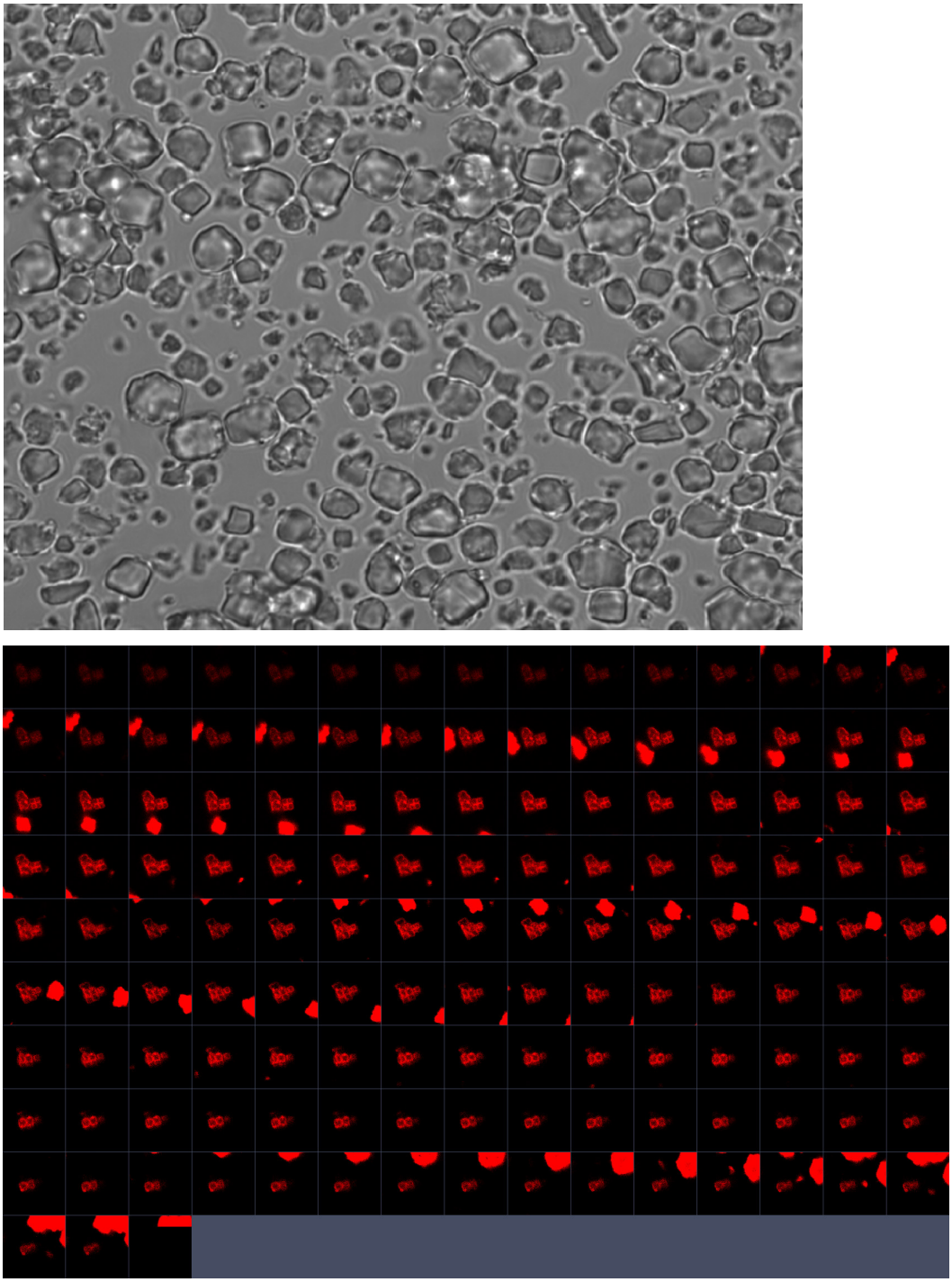

Figure S7. Confocal images of cubic $(PVPON/PMAA)_4$ microcapsules assemblies in buffer pH 4.5 C=0.01 M NaCl (top) and C=1 M NaCl (bottom). Cross sections of three dimensional (3D) assemblies of cubic $(PVPON/PMAA)_4$ microcapsules in buffer pH 4.5, C=1M at sequential time lapses (bottom).

[1] Good, R. J. *Contact Angle, Wettability and Adhesion; K.L. Mittal*, ed.; V.S.P. Utrecht: The Netherlands, 1993; pp. 3-36.

[2] Lee, L. H. in *Preprints, Div. Organic Coatings and Plastics Chemistry, 154th National Meeting of ACS*, Chicago, IL, 1967, p. 61

[3] Wu, S. *Org. Coat. Plast. Chem.*, 1971, **31**, 27.