Electronic Supplementary Material (ESI) for Nanoscale Advances.

Supplementary Information

Reconfigurable Assembly of Charged Polymer-Modified Janus and Non-Janus Particles: From Half-Raspberries to Colloidal Clusters and Chains

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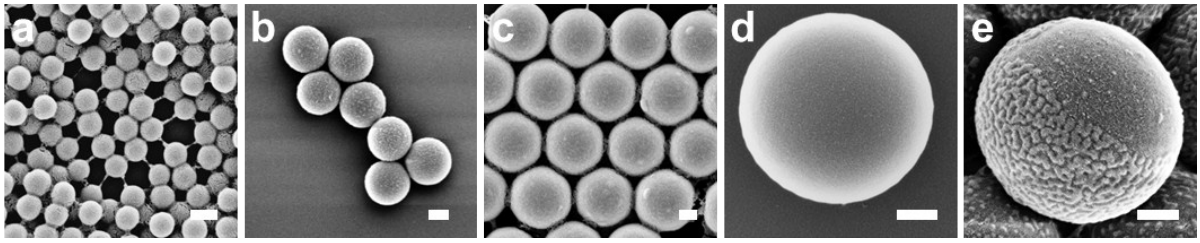

Figure S1. Representative SEM images of (a) 200-PAA-HP, (b) 450-PAA-HP, (c) 650-PAA-HP, (d) 1000-PDMAEMA-HP, and (e) 1000-PDMAEMA/PNIPAM-JP. Scale bar: 200 nm.

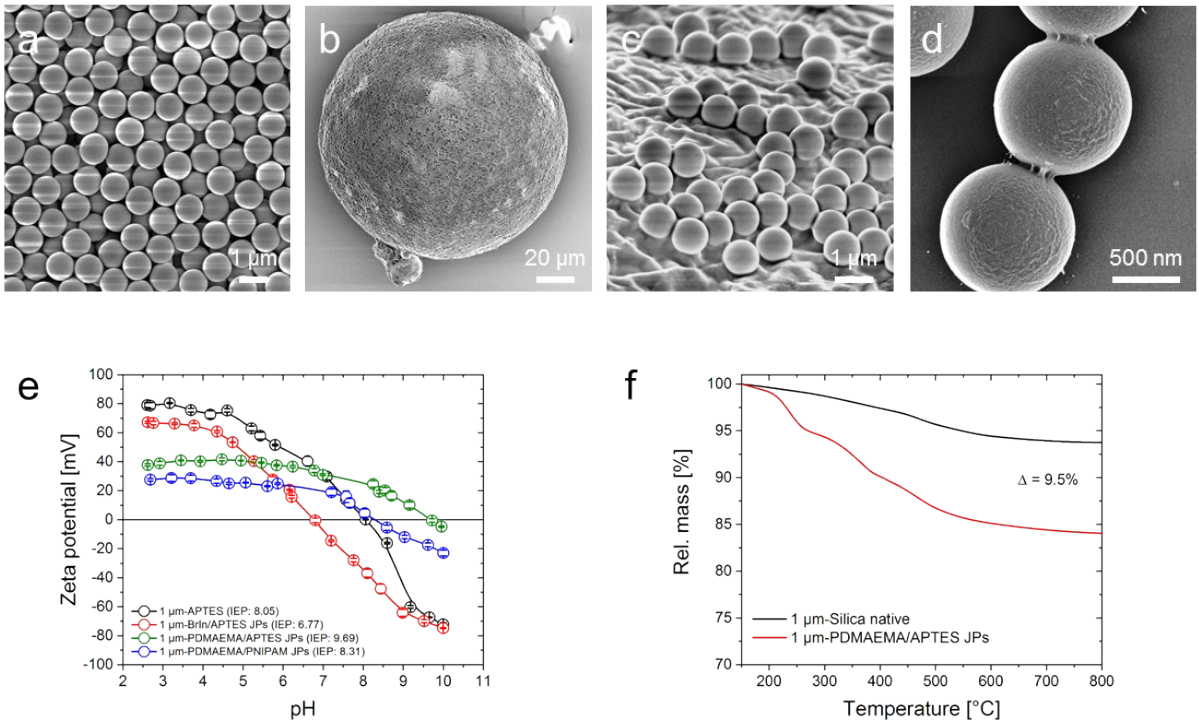

Figure S2. (a-d) Representative SEM images of APTES-modified 1 µm-silica particles (a), a colloidosomes (b), 1 µm APTES-modified silica particles immobilized at the wax-water interface (c), and 1 µm-PDMAEMA/APTES JPs (d). (e) Zeta potential vs. pH of premodified and JPs. (f) Dependence of the relative particle mass on the temperature obtained using TGA for 1 µm native and 1 µm-PDMAEMA-APTES-JPs.

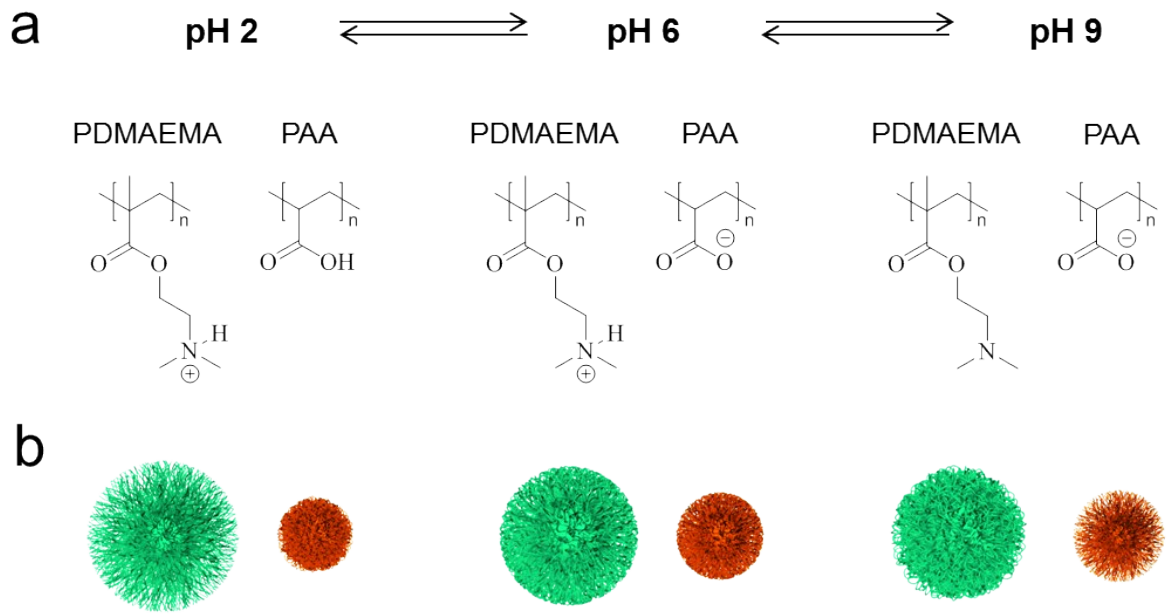

Figure S3. pH-responsive charges and swelling behavior of PDMAEMA (green) and PAA (red): (a) reversible pH-switchable charge state and (b) swelling state of the particle-covering polymer-brushes.

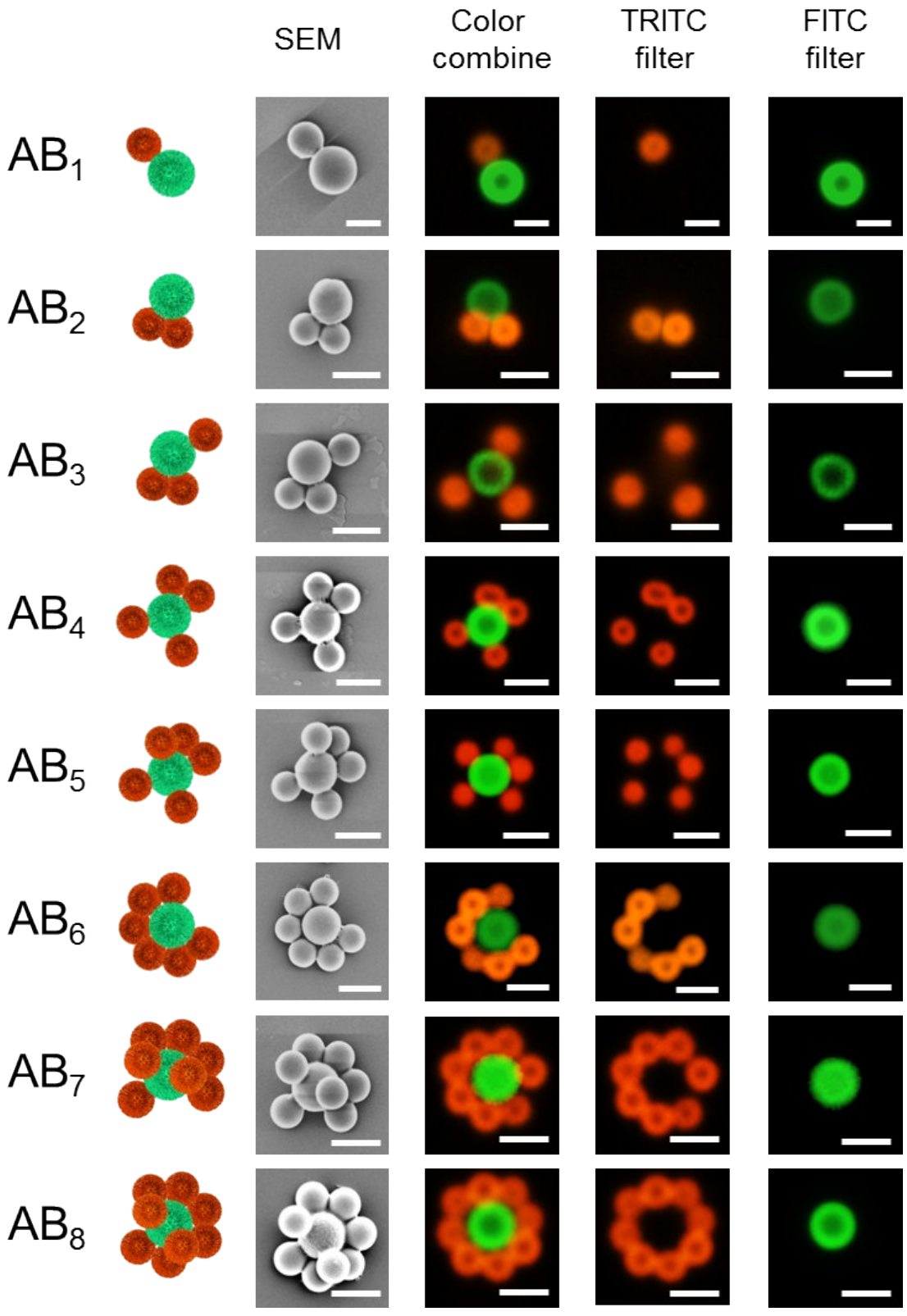

Figure S4. Observed raspberry-like structures: schematic and representative SEM and fluorescence microscopy images, formed in dispersion from homogeneously decorated 650-PAA-HP and 1000-PDMAEMA-HP (numerical ratio $N_{JP/PAA-HP}$ = 1:15). FLM images obtained with different filters and the combined colors. The ionic strength was adjusted to 1 mmol/l with KCl. Scale bars: 1 μm.

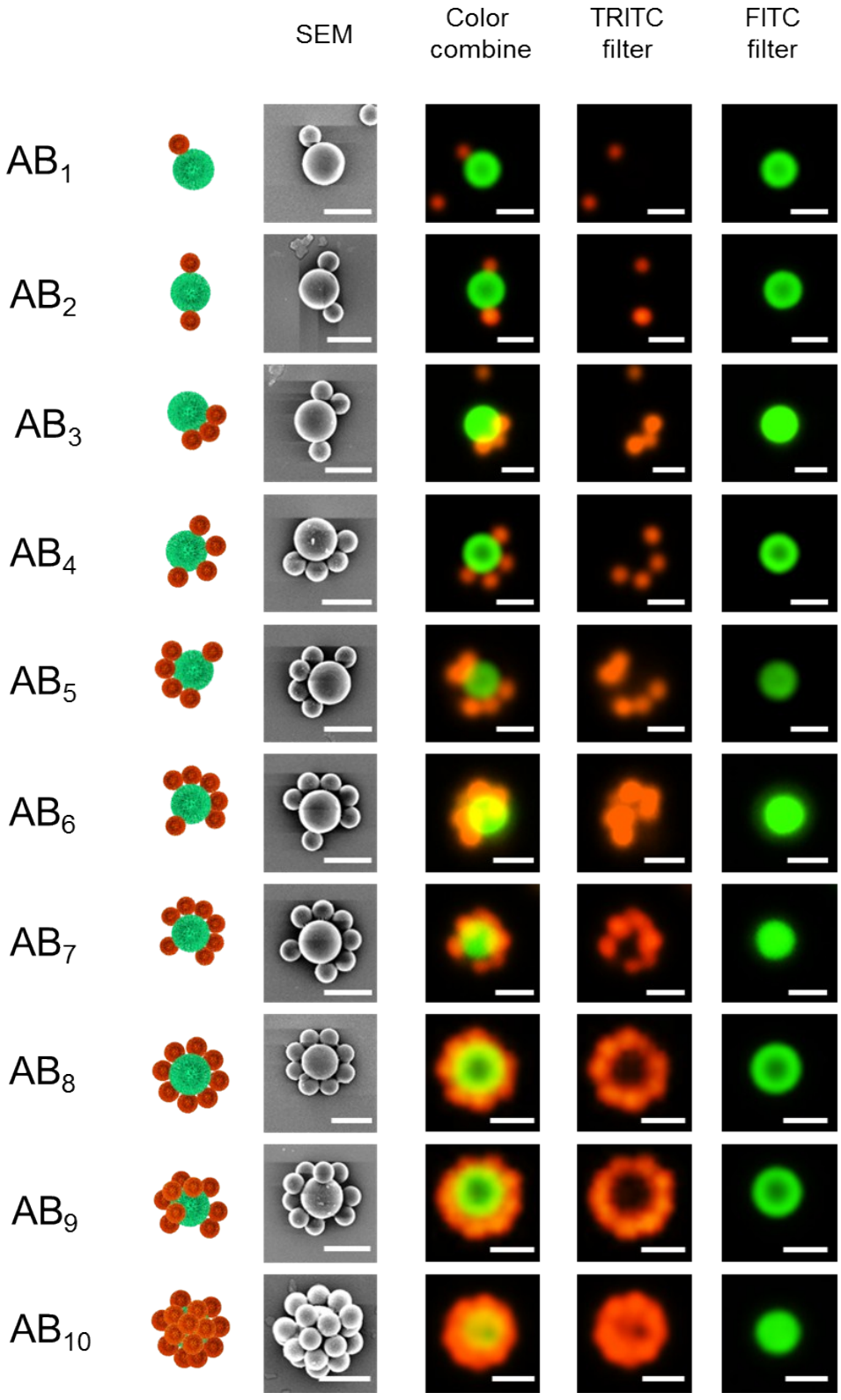

Figure S5. Observed raspberry-like structures: schematic and representative SEM and fluorescence microscopy images, formed in dispersion from homogeneously decorated 450-PAA-HP and 1000-PDMAEMA-HP (numerical ratio $N_{JP/PAA-HP}$ = 1:44). FLM images obtained with different filters and the combined colors. The ionic strength was adjusted to 1 mmol/l with KCl. Scale bars: 1 µm.

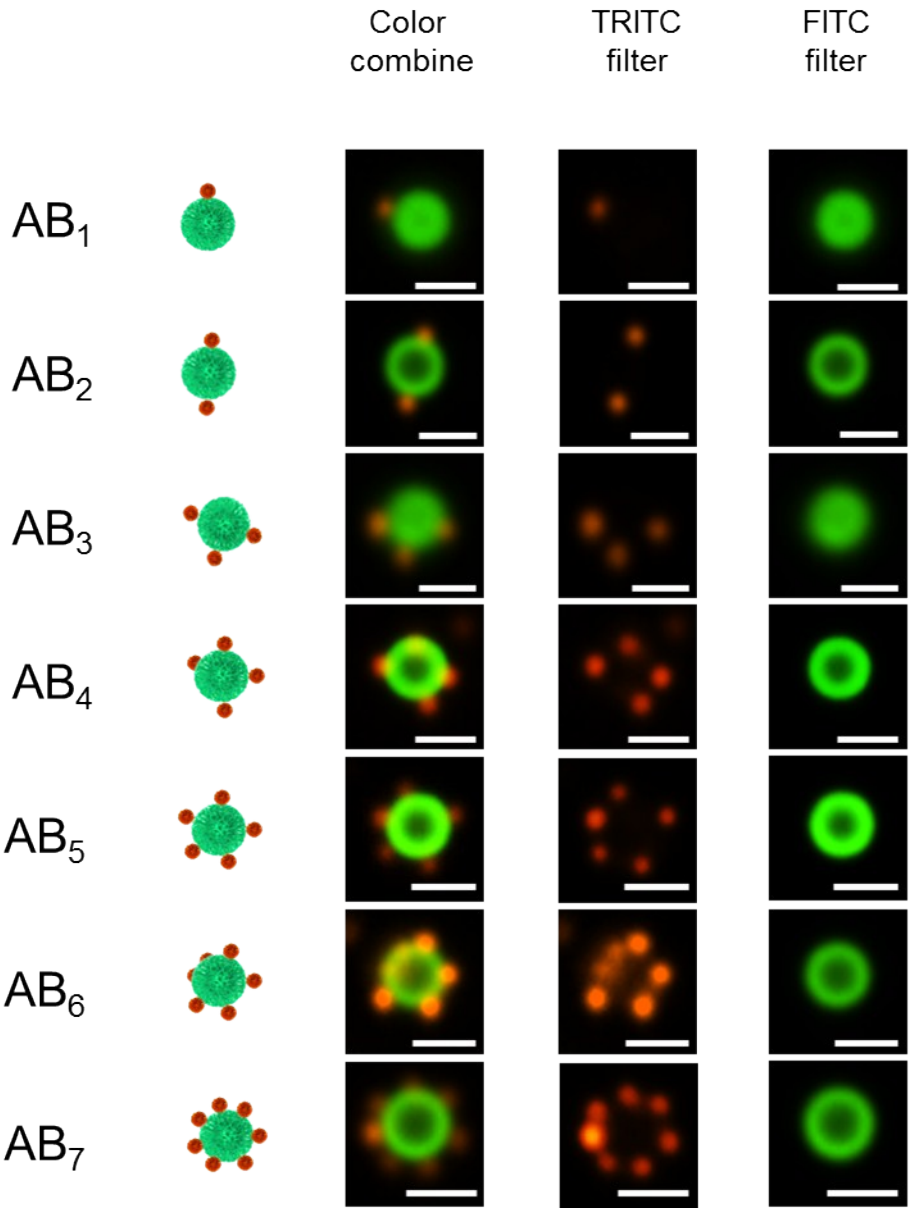

Figure S6. Observed raspberry-like structures: schematic and representative fluorescence microscopy images, formed in dispersion from homogeneously decorated 200-PAA-HP and 1000-PDMAEMA-HP (numerical ratio $N_{JP/PAA-HP}$ = 1:500). AB_8 to AB_{12}-type structures could be solely observed *in situ*, but could not be captured in 2D images, since the 3D arrangement of the 200-PAA-HP on the 1000-PDMAEMA-HP. Thus, some of the 200-PAA-HP are always out of focus when taking the image. FLM images obtained with different filters and the combined colors. The ionic strength was adjusted to 1 mmol/l with KCl. Scale bars: 1 μm.

Table S1: Average number of PAA-HP per JP for half-raspberry-like structures.

System	pH 6		pH 9		pH 6		pH 9		pH 6
200-PAA-HP	7.6		7.6		7.6		7.8		9.4
450-PAA-HP	8.9		8.0		7.6		6.9		6.6
650-PAA-HP	3.4	- 23%	2.6	+31%	3.4	-12%	3.0	+20%	3.6

Table S2: Average number of particles forming a cluster.

System	pH 6		pH 9		pH 6		pH 9		pH 6
200-PAA-HP	3.2	- 16%	2.7	+ 7%	2.9	- 3%	2.8	- 4%	2.7
450-PAA-HP	2.7	- 26%	2.0	+ 40%	2.8	- 29%	2.0	+ 30%	2.6
650-PAA-HP	2.9	+ 31%	3.8	- 8%	3.5	+ 3%	3.6	- 22%	2.8

Table S3: Average number of particles forming a chain.

System	pH 6		pH 9		pH 6		pH 9		pH 6
200-PAA-HP	4.0	- 5%	3.8	+ 5%	4.0	± 0%	4.0	± 0%	4.0
450-PAA-HP	3.6	- 11%	3.2	+ 16%	3.7	- 19%	3.0	+ 13%	3.4
650-PAA-HP	3.0	± 0%	3.0	+ 3%	3.1	± 0%	3.1	- 3%	3.0

Table S4: Average number of particles forming a cluster or chain.

System	pH 6		pH 9		pH 6		pH 9		pH 6
200-PAA-HP	3.3	- 12%	2.9	+ 10%	3.2	- 3%	3.1	+ 3%	3.2
450-PAA-HP	2.9	- 28%	2.1	+ 38%	2.9	- 28%	2.1	+ 29%	2.7
650-PAA-HP	3.2	+ 13%	3.6	- 6%	3.4	+ 6%	3.6	- 22%	2.8