Supplementary Information

Omnidispersible Microscale Colloids with Nanoscale Polymeric Spikes

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Table S1. Zinc content measured by energy dispersive spectroscopy (EDS) of TPs after different acid washes (72 h) to remove ZnO nanorods and after glutaraldehyde crosslinking.

Sample	Zinc Content (by EDS)	Acid Wash
0.57 ZnTP	0.57 ± 0.12	0.1 M HNO_3
2.9 ZnTP	2.9 ± 0.21	0.01 M HNO_3
43 ZnTP	43.0 ± 3.3	None

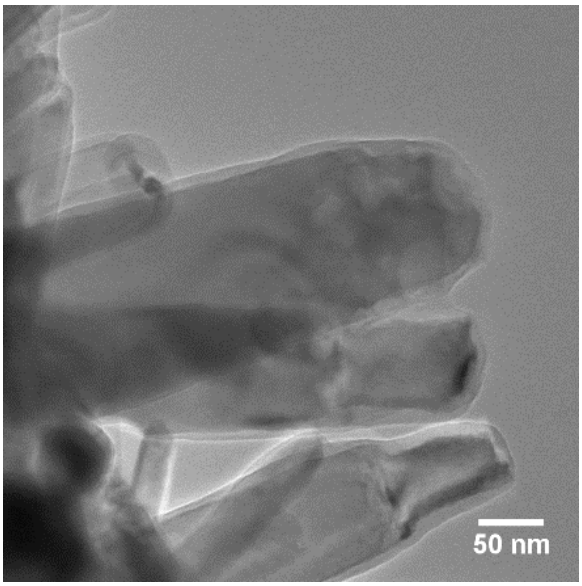

Figure S1: TEM image of a conformal $(PAA/PAH)_2$ bilayer film on a HP.

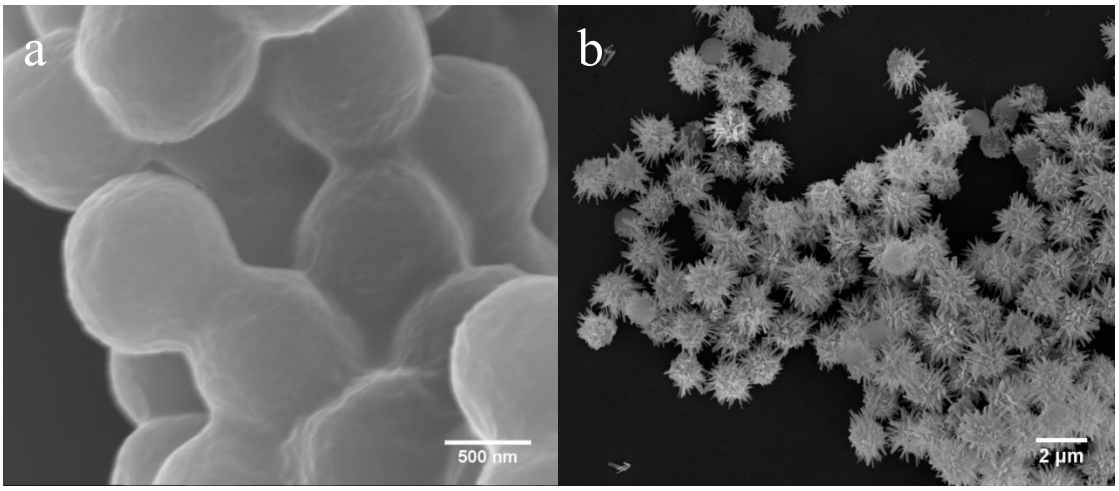

Figure S2: SEM of HPs modified with $(PAA/PAH)_2$ after pH 4 treatment with no glutaraldehyde for 2 hours (**a**) and normal HPs with no polymer film after being treated with glutaraldehyde (**b**).

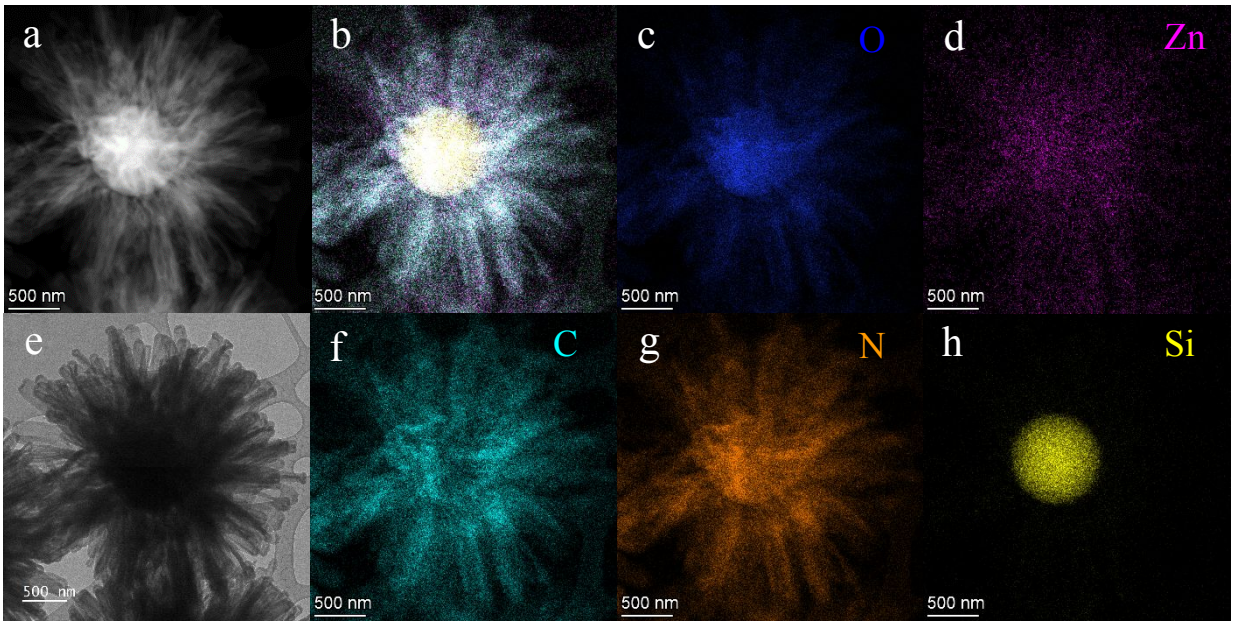

Figure S3: EDS map and electron microscopy images of a TP cross-linked with glutaraldehyde washed with 1M boric acid for 72 hours: (**a**) STEM HAADF image; (**b**) composite EDS map; (**c**) oxygen EDS map; (**d**) zinc EDS map; (**e**) TEM image; (**f**) carbon EDS map; (**g**) nitrogen map; (**h**) silicon map.

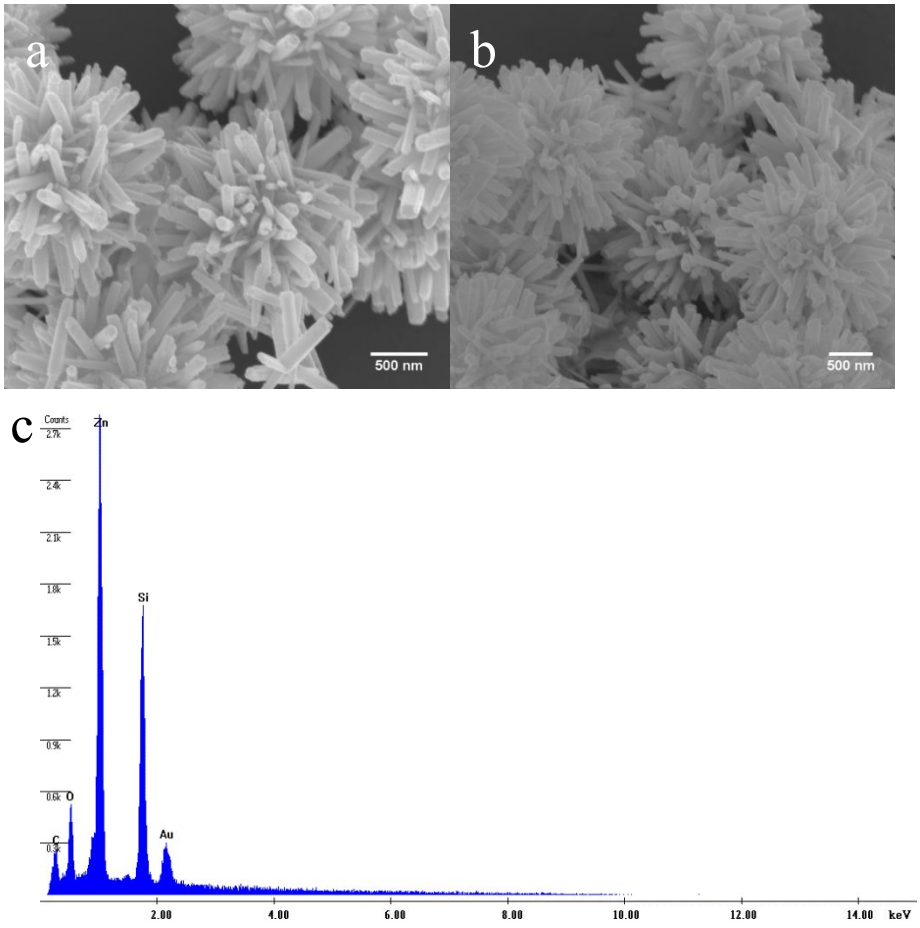

Figure S4: TPs formed with glutaraldehyde cross-linking at pH 10 (**a**) and pH 7 (**b**). Representative EDX spectra from glutaraldehyde at high pH (**c**).

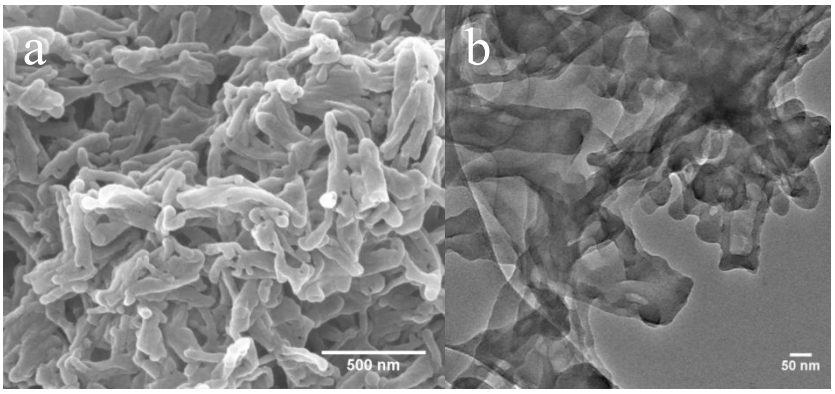

Figure S5: SEM (**a**) and TEM images (**b**) of ZnO NRs coated with $(PAA/PAH)_2$ and cross-linked with glutaraldehyde at pH 4 in an analogous process to creation of TPs.

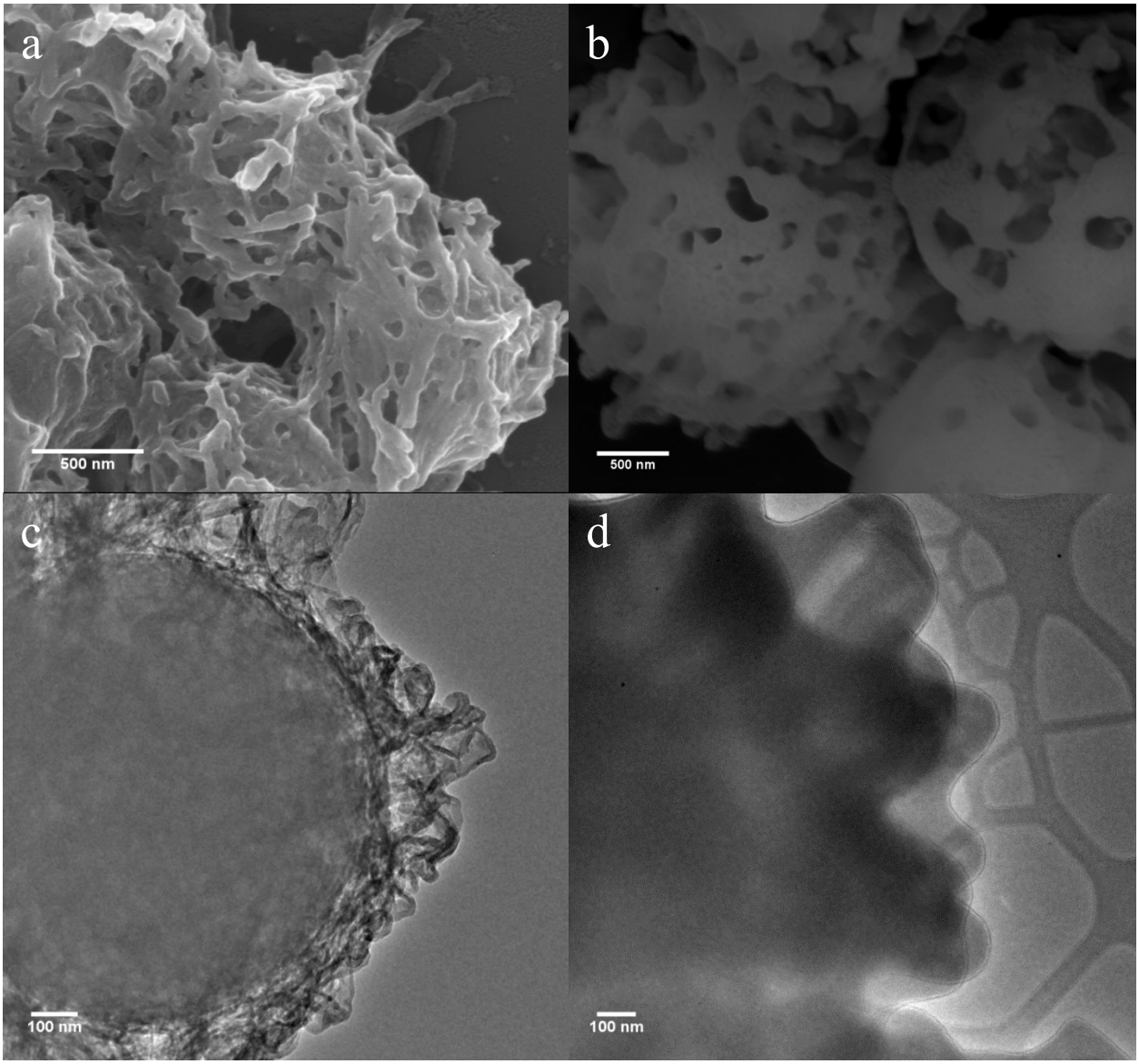

Figure S6: SEM (**a,b**) and TEM (**c,d**) images of a polystyrene crosslinked 2 bilayer $(PAA/PAH)_2$ TP made from a HPs with 64 nm diameter spikes of 445 nm length (**a,c**) and a cross-linked five-bilayer PAA/PAH film (**c,d**).

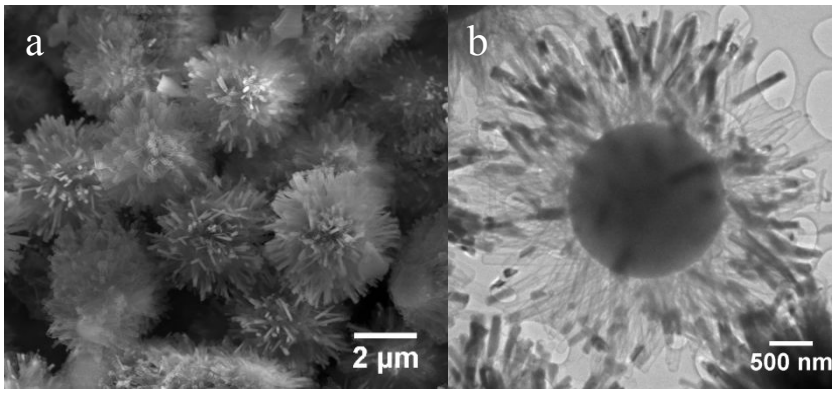

Figure S7: SEM (**a**) and TEM (**b**) of TPs made from HPs with an average spike length of 1930 nm and average spike width of 146 nm after glutaraldehyde crosslinking and a shorter 4 hour treatment with 1 M boric acid of HP.

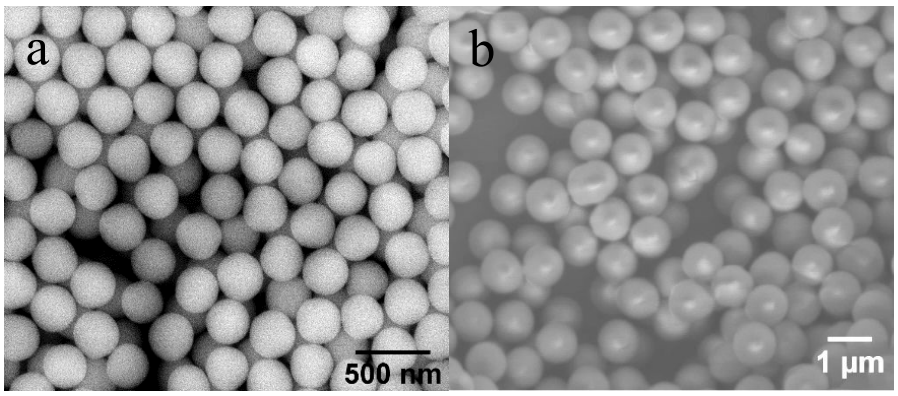

Figure S8: SEM image of (**a**) 296 nm average diameter SiO_2 beads and (**b**) 1.1 µm average diameter SiO_2 particles producing using Stöber method and modified Stöber method respectively.

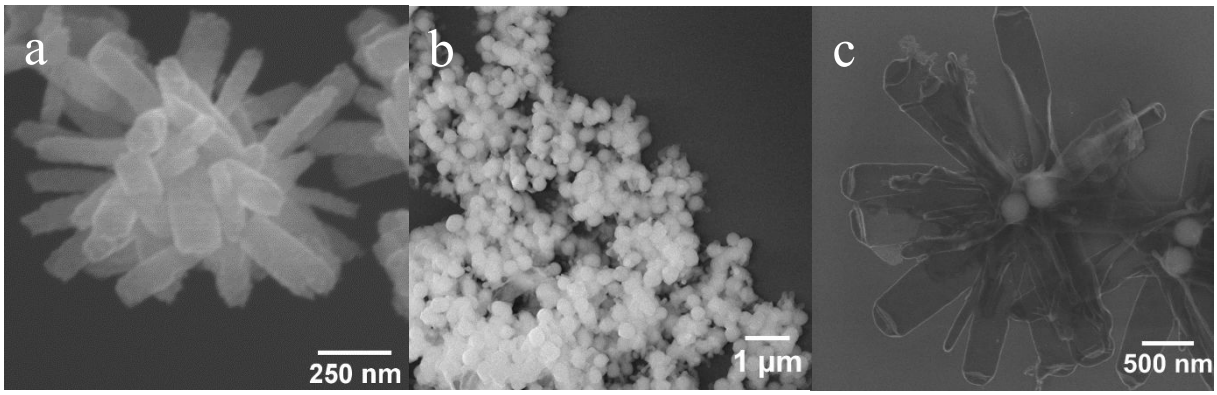

Figure S9: (**a**) SEM image of HP made from 296 nm average diameter SiO_2 cores with spikes of ca. 98 nm width and ca. 411 nm length. (**b**) TEM image of corresponding TPs formed from (**a**). (**c**) SEM image of TPs made from HPs with 296 nm average diameter SiO_2 cores with average spike length of 1253 nm and average spike width of 188 nm.

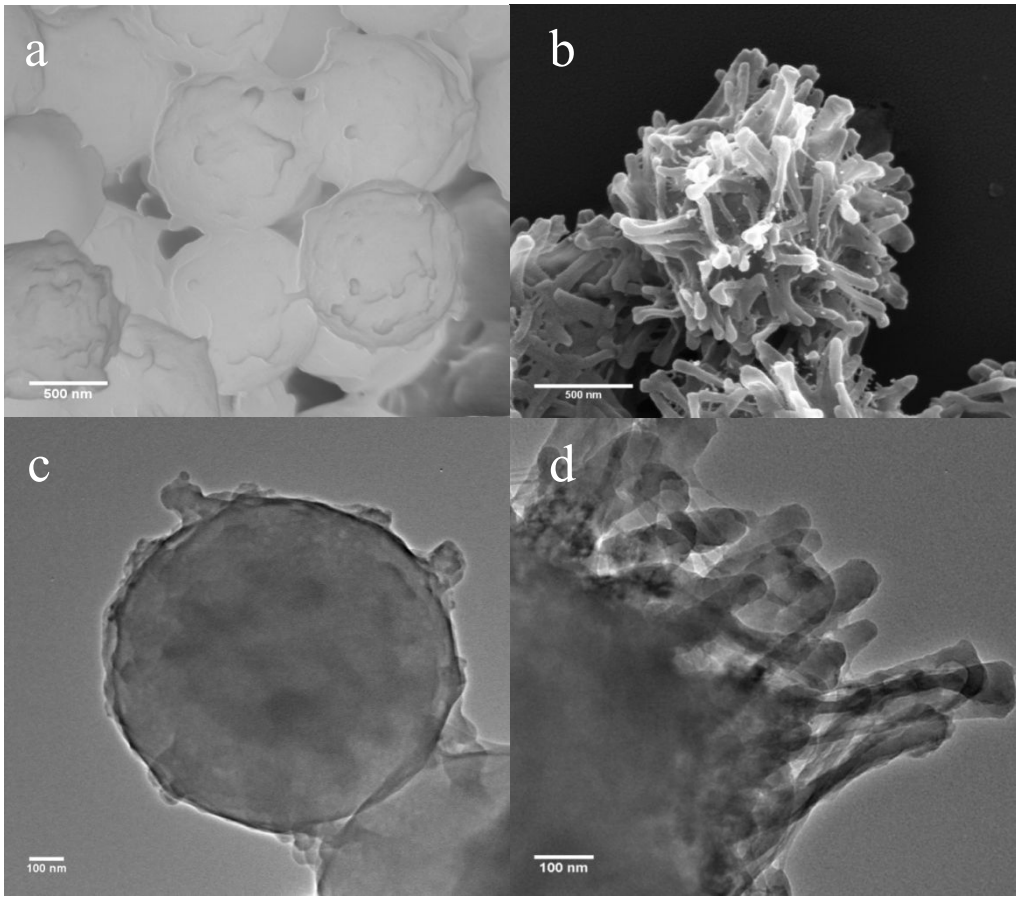

Figure S10: SEM (**a,b**) and TEM (**c,d**) images of $(PAA/PAH)_2$ TP (**a,c**) and $(PAA/PAH)_3PAA$ TP (**b,d**) cross-linked with EDC.

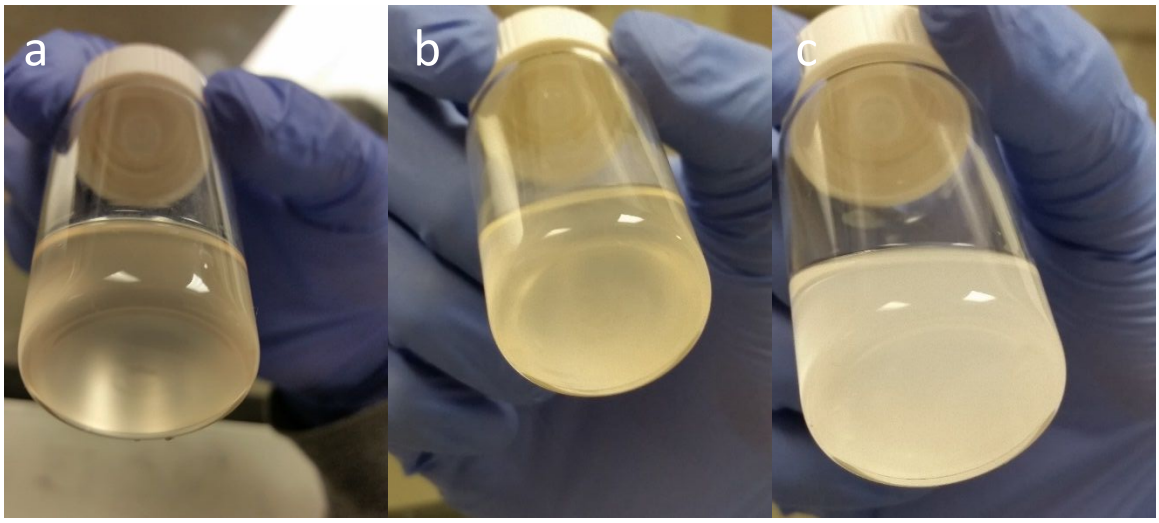

Figure S11: Photographs of dispersions of 0.57-Zn (**a**), 2.9-Zn (**b**), and 43-Zn TPs (**c**) in heptane (0.5 mg/mL).

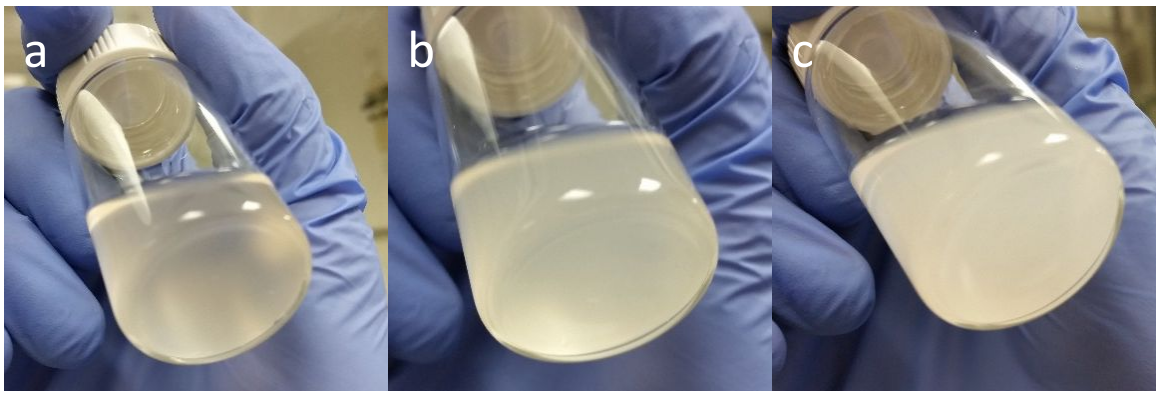

Figure S12: Photographs of dispersions of 0.57-Zn (**a**), 2.9-Zn (**b**), and 43-Zn TPs (**c**) in 1 M NaCl (0.5 mg/mL)

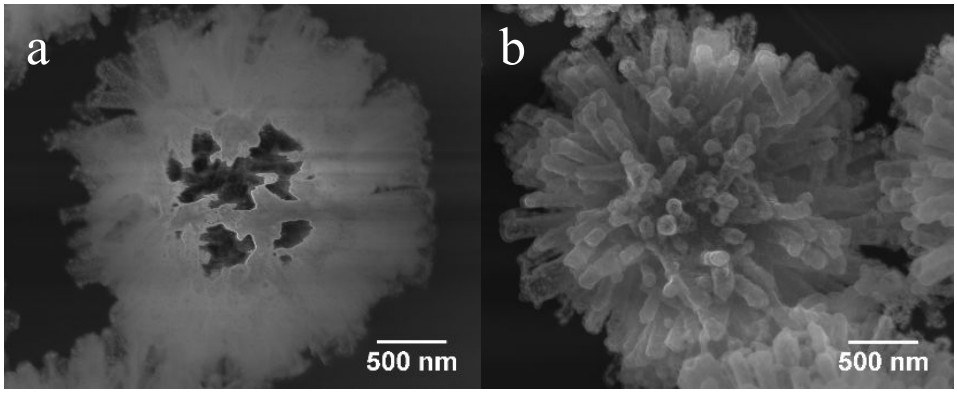

Figure S13: SEM images of TPs with PSS/PDDA/Au NP (Au TP) (**a**) and with PSS/PDDA (0.5 M NaCl)/AuNP (AuSTP) layers encapsulated (**b**).

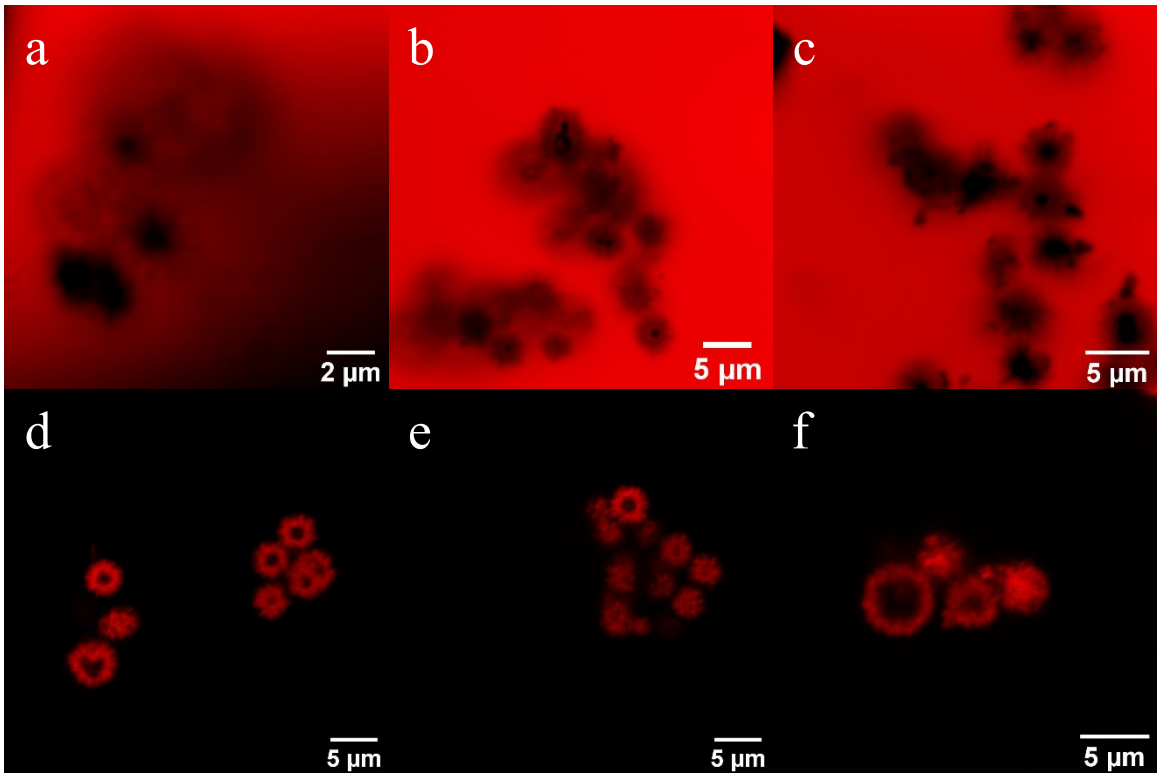

Figure S14: 640 nm confocal microscopy of Nile Red (NR) in ethanol (a), 1 M Rhodamine B (RhB) at pH 3, (b) 1 M RhB at pH 10, (c) encapsulation of NR at pH 3, (d) encapsulation of NR at pH 10, and (e) encapsulation RhB at pH 10. All experiments were done with TPs at 0.1 mg/mL. Encapsulation involved repeated washing at the appropriate pH to remove any excess dye after incubation in 1 mM in ethanol (NR) or in pH 3 water (RhB).

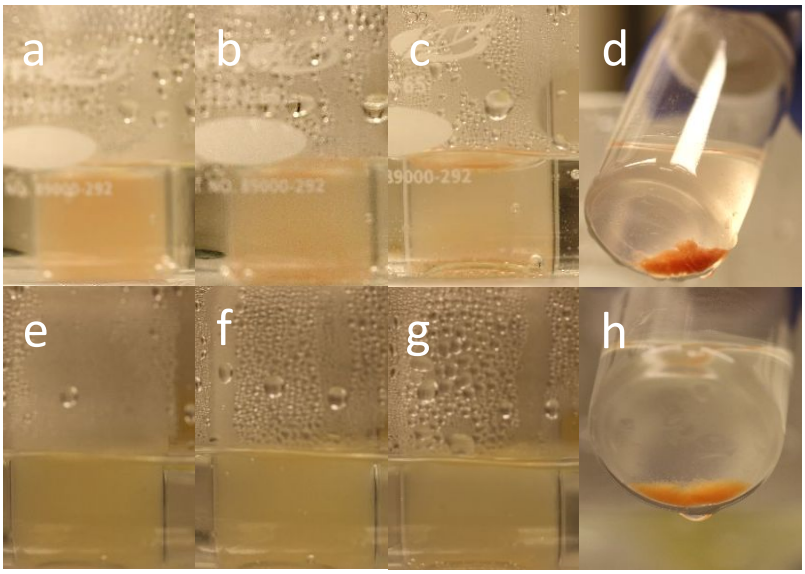

Figure S15: Photographs of dispersions of PNIPAM TPs (**a-d**) and PAA TPs (**e-h**) at 80 °C after (**a,e**) 1 minute (**b,f**) 5 minutes (**c,g**) 10 minutes. Sediments after being incubated in a water bath for 10 minutes at 80 °C (**d,h**).

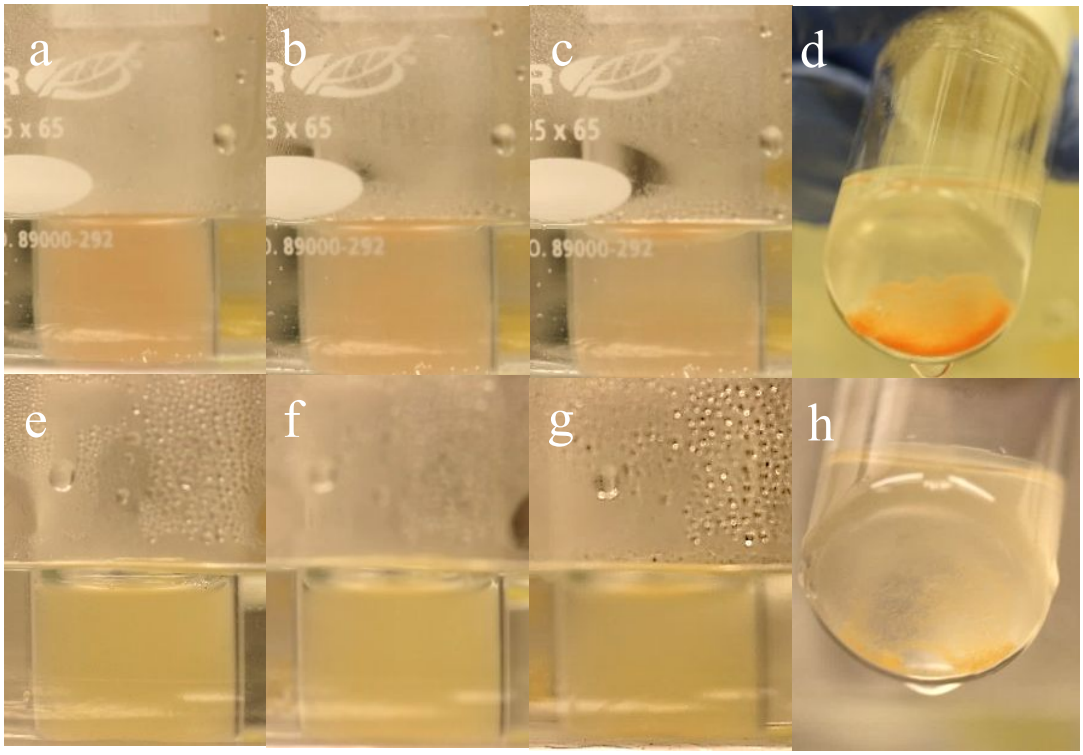

Figure S16: Photographs of dispersions of PNIPAM TPs (**a-d**) and PAA TPs (**e-h**) at 60 °C after (**a,e**) 1 minute (**b,f**)5 minutes (**c,g**) 10 minutes. Sediments after being incubated in a water bath for 10 minutes at 60 °C (**d,h**).

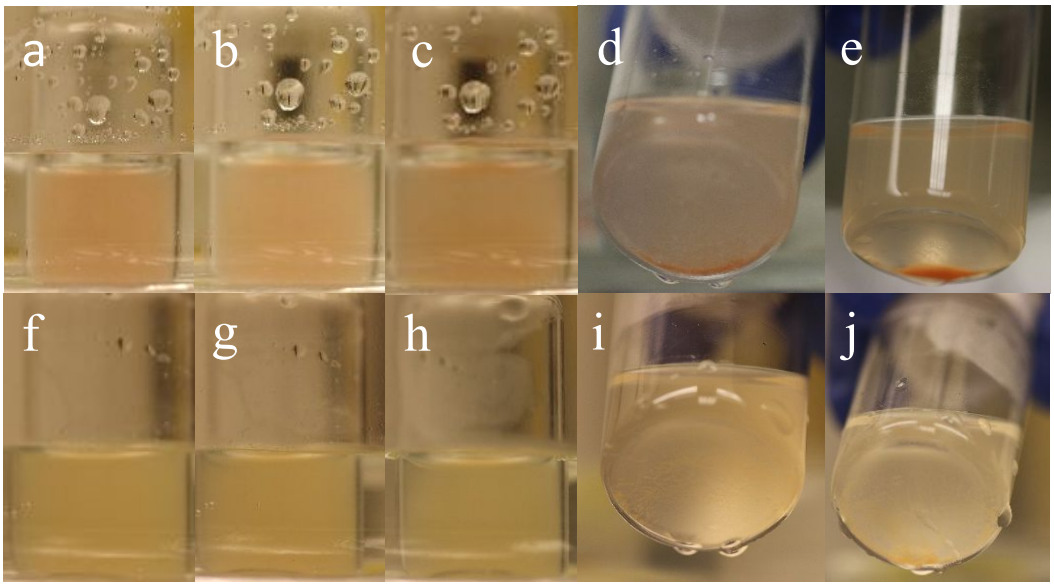

Figure S17: Photographs of dispersions of PNIPAM TPs (**a-e**) and PAA TPs (**f-j**) at 40 °C after (**a,f**) 1 minute (**b,g**) 5 minutes (**c,h**) 10 minutes. Sediments after being incubated in a water bath for 5 minutes (**d,i**) and 10 minutes (**e,j**) at 40 °C.

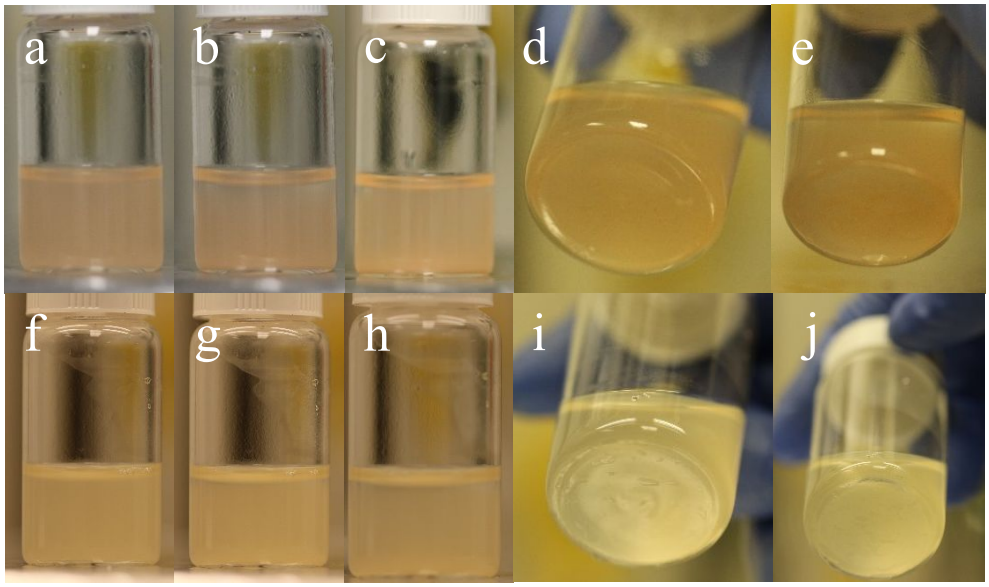

Figure S18: Photographs of dispersions of PNIPAM TPs (**a-e**) and PAA TPs (**f-j**) at room temperature after (**a,f**) 1 minute (**b,g**) 5 minutes (**c,h**) 10 minutes. Sediments after 5 minutes (**d,i**) and 10 minutes (**e,j**) at room temperature.

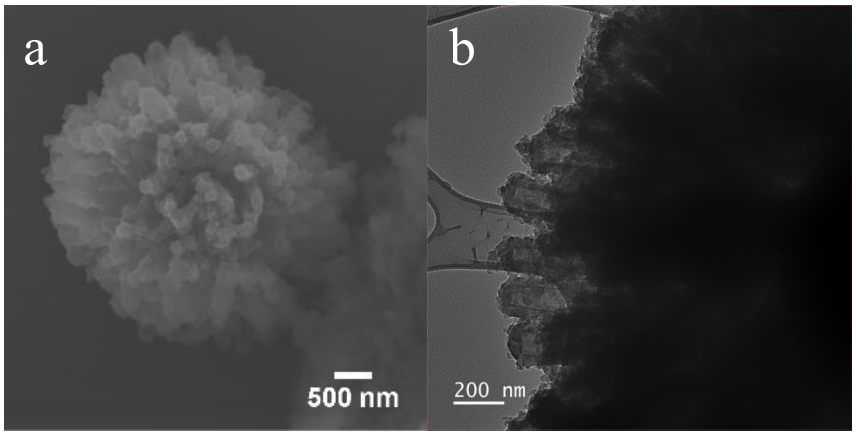

Figure S19: SEM (**a**) and TEM images (**b**) of dopamine-modified TPs (DOP TPs).

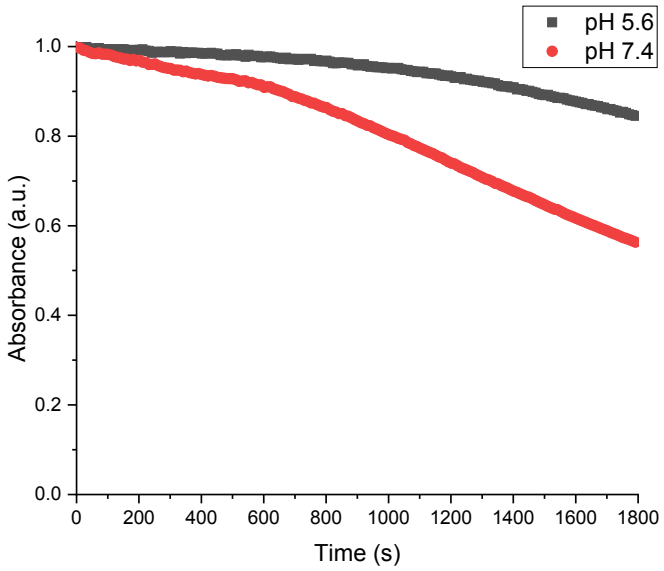

Figure S20: Absorbance measurements of Dopamine-modified TPs (DOP TPs) at pH 5.6 and pH 7.4 in 0.01 M PBS buffer in a quartz cuvette taken at wavelength of 275 nm every 15 seconds for 15 minutes.

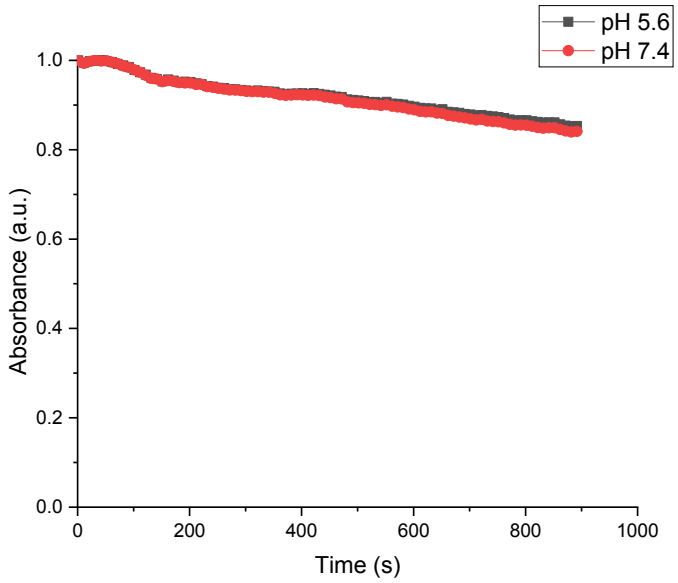

Figure S21: Absorbance measurements of PAA TPs at pH 5.6 and pH 7.4 in .01 M PBS buffer taken at wavelength 275 nm every 15 seconds for 15 minutes.

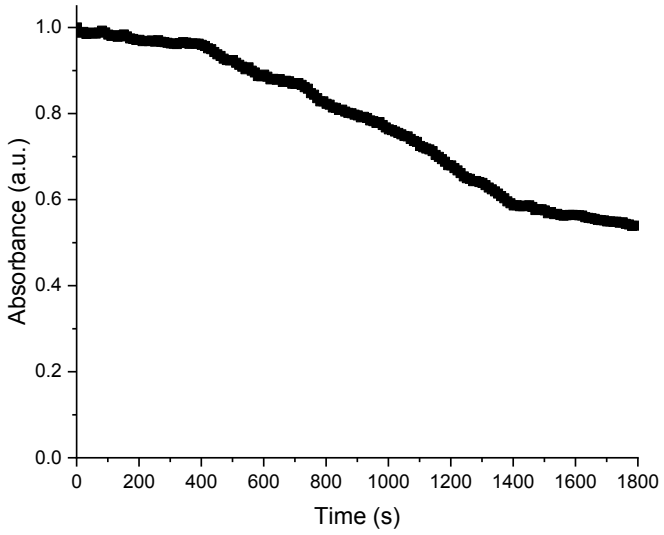

Figure S22: Absorbance measurements of DOP TPs at pH 5.6 in 01 M PBS buffer after previously being adjusted to pH 7.4 in 01 M PBS buffer for 30 minutes taken at 275 nm absorbance band every 15 seconds for 30 minutes.