

## *Supporting Information*

# **Double in situ Preparation of Raspberry-like Polymer Particles**

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## List of Contents

Table S1. Summary of formulations, particle sizes and PDIs for the PSt-SiO <sub>2</sub> NCPs prepared by the postpolymerization method	S3
Figure S1. Size distribution of raspberry-like PSt-SiO <sub>2</sub> NCPs determined by DLS (see Table 1, runs 2 and 4)	S4
Figure S2. TEM images of the as-obtained PSt-SiO <sub>2</sub> NCPs using SDS as stabilizer	S4
Figure S3. (a) TEM image and (b) DLS size distribution of PSt-SiO <sub>2</sub> NCPs (see Table 1, run 12)	S5
Figure S4. TEM image of the dried emulsion from run S2 (see Table S1) after the formation of SiO <sub>2</sub> nanoparticles for 3 d at room temperature (RT) before the polymerization	S5
Figure S5. TEM images of the PSt-SiO <sub>2</sub> NCPs prepared by the postpolymerization method with different mass ratios St/TEOS and amounts of 4-VP	S6
Figure S6. DLS size distribution of PSt-SiO <sub>2</sub> NCPs (see Table S1, run S5)	S7
Figure S7. TEM micrograph (a) and EDX element mappings of C (b) and Si (c) of PSt-SiO <sub>2</sub> NCPs at a polymerization time of 5 h with an initial ratio of St/TEOS of 70/30 and 25 mg of 4-VP (Table 1, run 2)	S7
Figure S8. TEM images of SiO <sub>2</sub> particles after different times of the sol-gel process from emulsions with TEOS and toluene as dispersed phase	S8

**Table S1.** Summary of formulations, particle sizes and PDIs for the PSt-SiO<sub>2</sub> NCPs prepared by the postpolymerization method using the waterborne initiator (V-057) and Eosin Y as stabilizer (2 mg/mL).

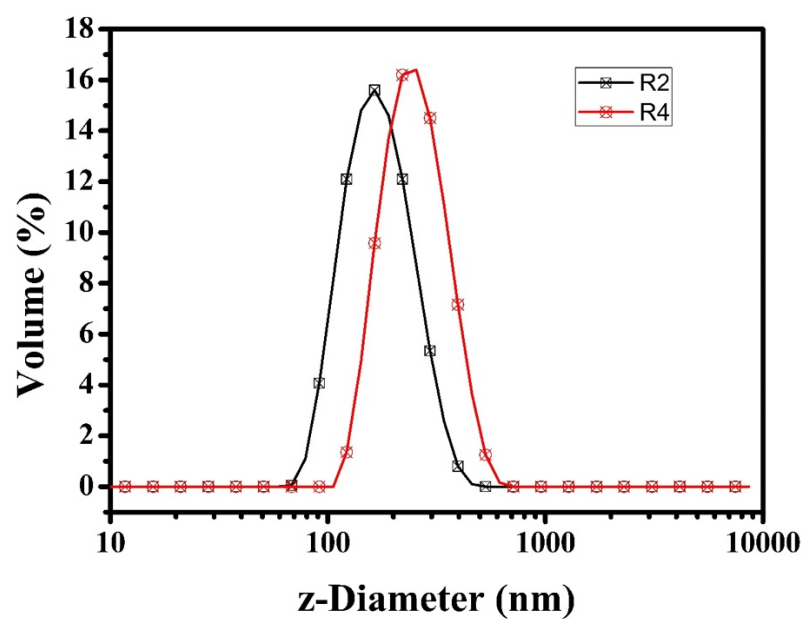
Runs	St/g	TEOS/g	4-VP/mg <sup>a</sup>	HD/mg	Initiator/mg	Reaction conditions	z-Diameter/nm ( $D_z$ ) <sup>b</sup>	Polydispersity (PDI) <sup>b</sup>
S1 <sup>c</sup>	2	0	0	80	80	60 °C, 5h	239	0.083
S2	0.45	0.05	50	20	20	RT, pH 9, 3 d; <sup>d</sup> 60 °C, 5 h	114	0.042
S3	0.45	0.05	75	20	20	RT, pH 9, 3 d; <sup>d</sup> 60 °C, 5 h	148	0.028
S4	0.4	0.1	75	20	20	RT, pH 9, 3 d; <sup>d</sup> 60 °C, 5 h	157	0.136
S5	0.35	0.15	75	20	20	RT, pH 9, 3 d; <sup>d</sup> 60 °C, 5 h	132	0.098

<sup>a</sup> After the formation of SiO<sub>2</sub> nanoparticles for 3 d at room temperature (RT), a certain mass fraction of 4-VP was added into the emulsion.

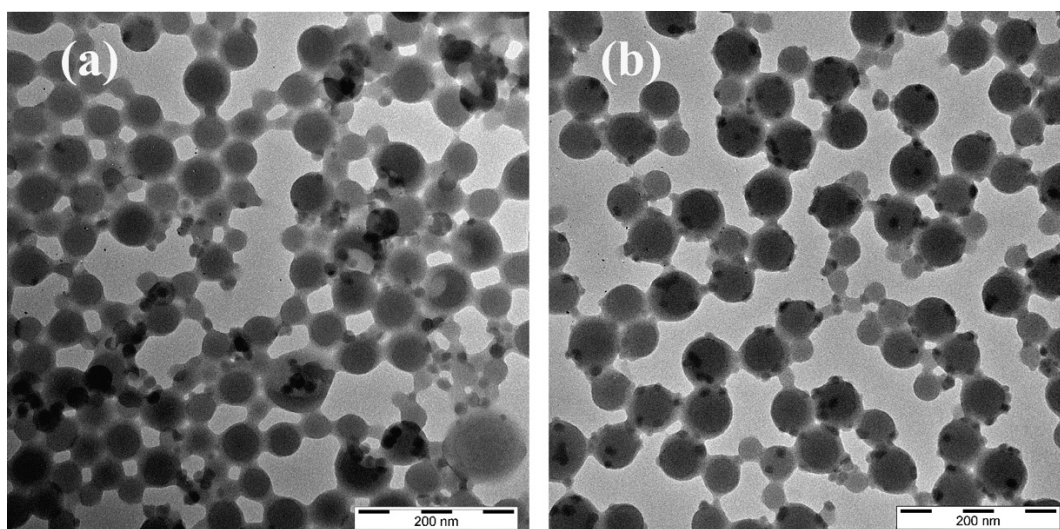
<sup>b</sup> Determined by DLS.

<sup>c</sup> The monomer concentration is 20 wt%.

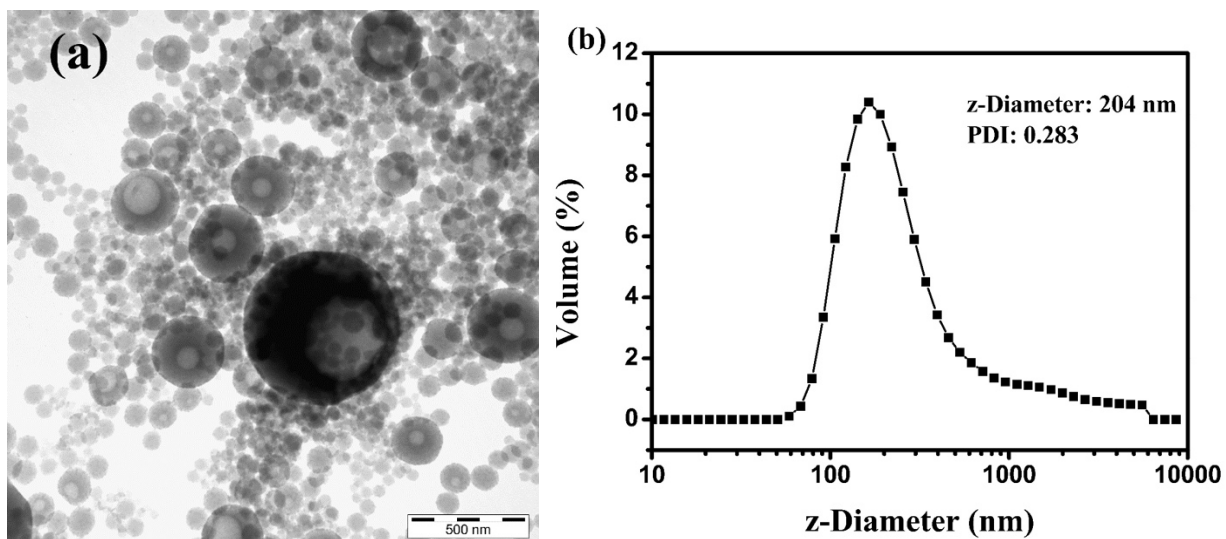
<sup>d</sup> In a 0.01 M NH<sub>3</sub>\*NH<sub>4</sub>Cl buffered aqueous solution at pH = 9.



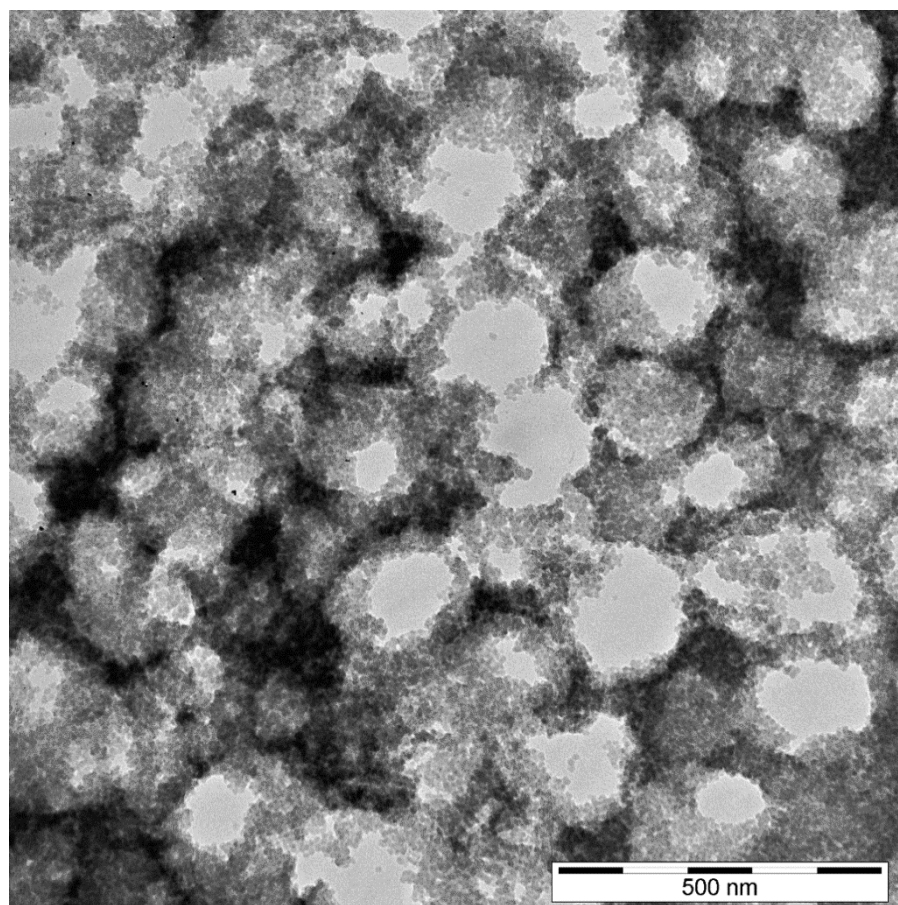
**Figure S1.** DLS size distribution of raspberry-like PSt-SiO<sub>2</sub> NCPs (see Table 1, runs 2 and 4).



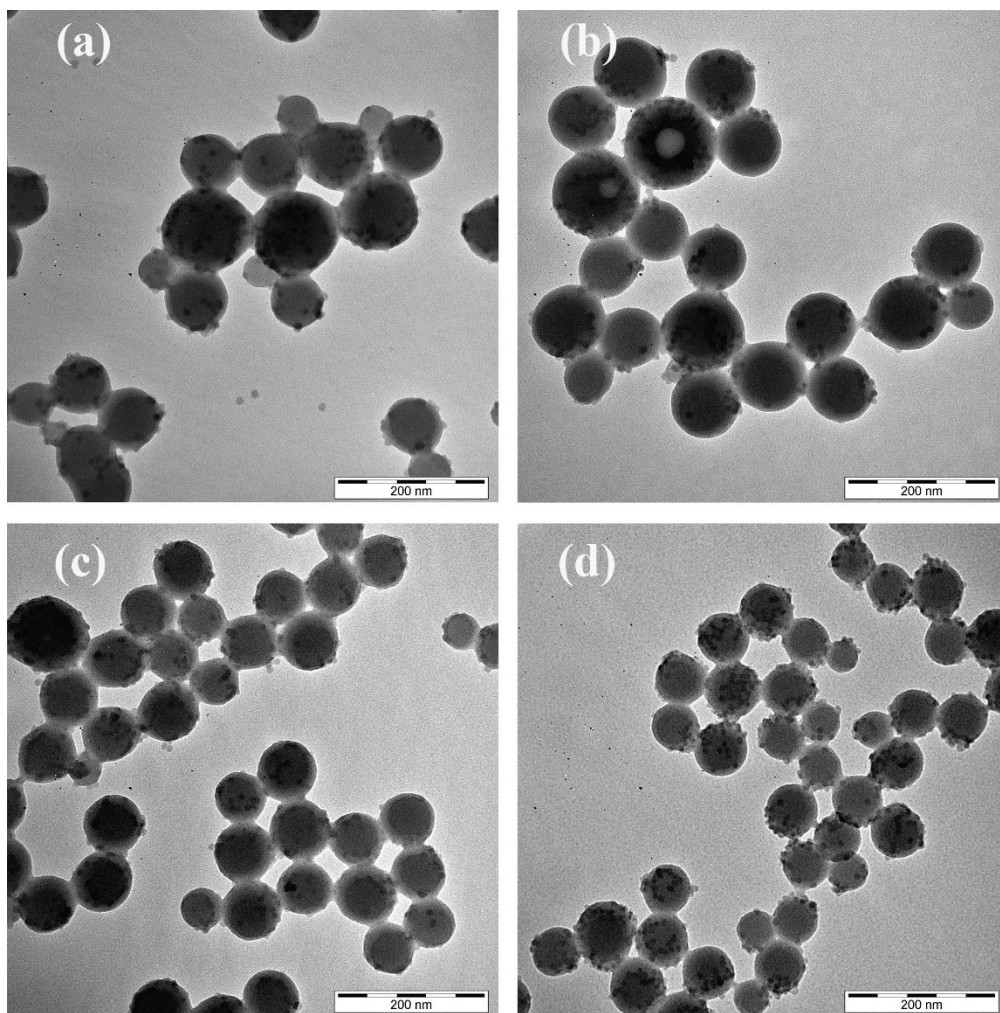
**Figure S2.** TEM images of the as-obtained PSt-SiO<sub>2</sub> NCPs prepared using SDS as stabilizer (see Table 1, runs 9 and 10).



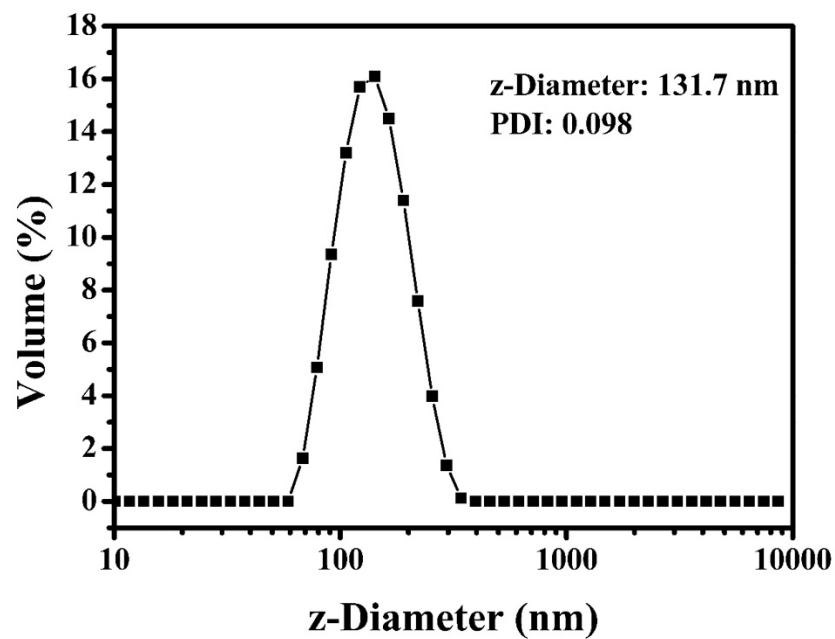
**Figure S3.** (a) TEM image and (b) DLS size distribution of PSt-SiO<sub>2</sub> NCPs (see Table 1, run 12).



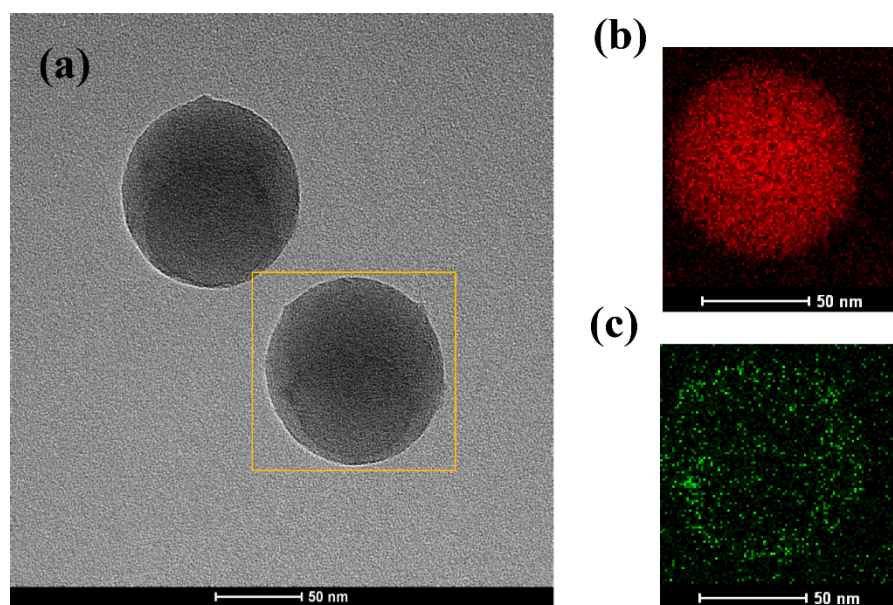
**Figure S4.** TEM image of the dried emulsion from run S2 (see Table S1) after the formation of SiO<sub>2</sub> nanoparticles for 3 d at room temperature (RT) before the polymerization.



**Figure S5.** TEM images of the PSt-SiO<sub>2</sub> NCPs prepared by the postpolymerization method with different mass ratios St/TEOS and amounts of 4-VP: (a) 90/10 St/TEOS, 50 mg of 4-VP, (b) 90/10 St/TEOS, 75 mg of 4-VP, (c) 80/20 St/TEOS, 75 mg of 4-VP and (d) 70/30 St/TEOS, 75 mg of 4-VP (see Table S1, runs S2-S5).

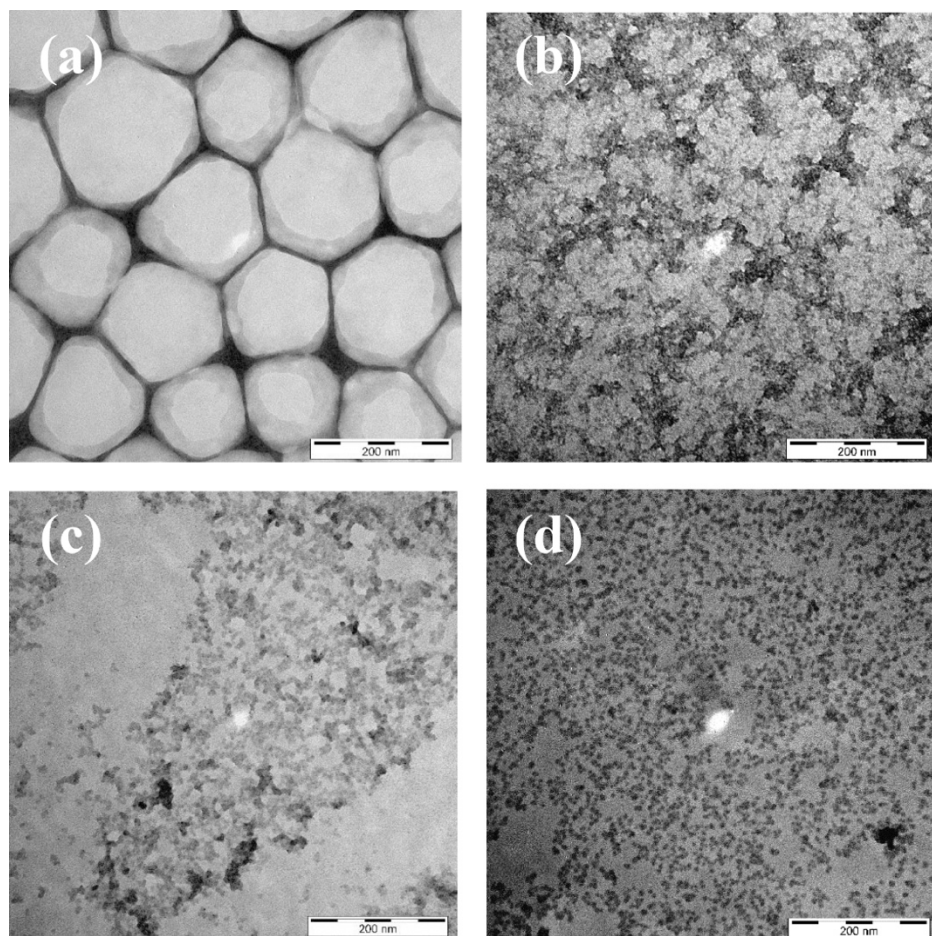


**Figure S6.** DLS size distribution of PSt-SiO<sub>2</sub> NCPs (see Table S1, run S5).



**Figure S7.** TEM micrograph (a) and EDX element mappings of C (b) and Si (c) of PSt-SiO<sub>2</sub> NCPs at a polymerization time of 5 h with an initial ratio of St/TEOS of 70/30 and 25 mg of 4-VP (Table 1, run 2).





**Figure S8.** TEM images of SiO<sub>2</sub> particles after different times of the sol-gel process from emulsions with TEOS and toluene as dispersed phase: (a) 5 h, (b) 12 h, (c) 18 h and (d) 24 h.