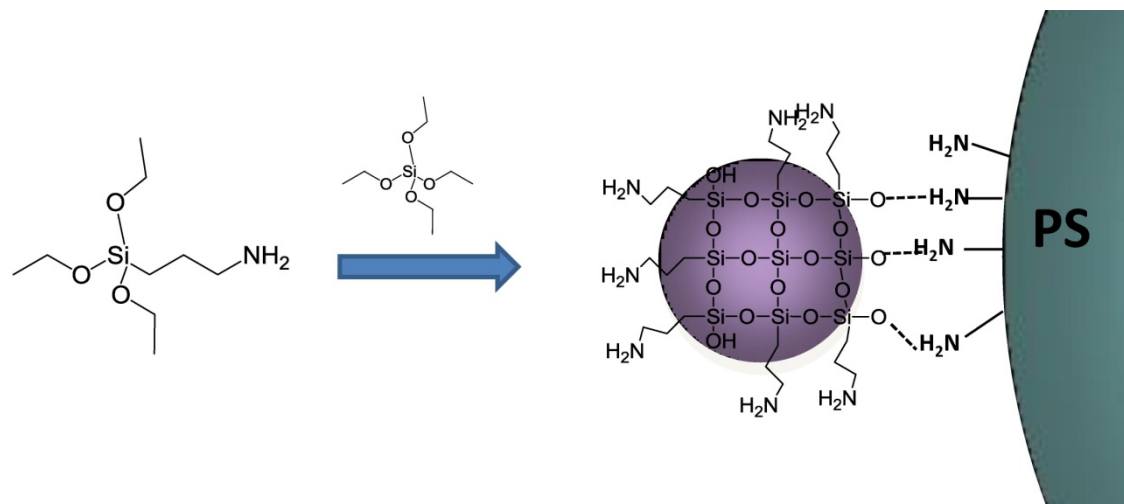


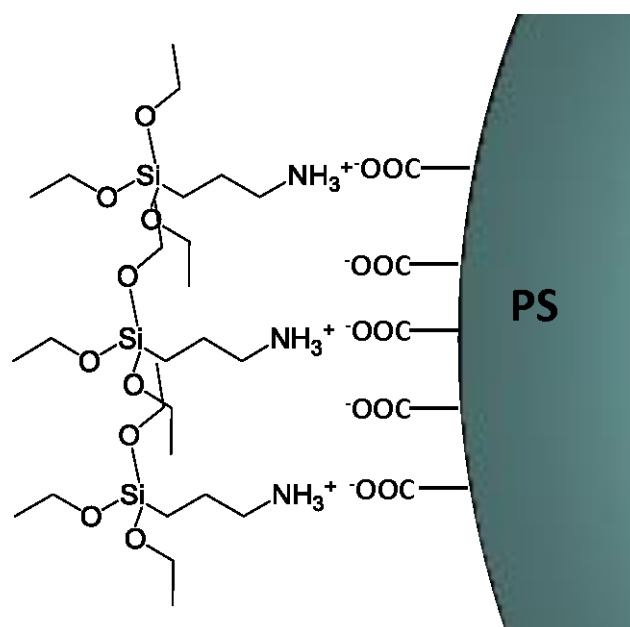
## Supporting Information

One-pot synthesis of surface roughness controlled hollow silica spheres with enhanced drug loading and release profile under ambient conditions in aqueous solutions

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Scheme S1 Schematic illustration showing the formation of spherical silica nuclei due to the pre-adsorption and hydrolysis of APTES and TEOS and their interactions with amino-PS particles. The nucleated tiny silica particles were attached to amino-PS particle surfaces probably through hydrogen bonds. Further addition of TEOS will lead to silica growth on the attached nuclei and formation of raspberry-like structure (Scheme 1).



Scheme S2 Schematic illustration showing that the electrostatic interactions between APTES and carboxyl-PS particles resulted in the formation of a monolayer of cross-linked APTES directly attached on the negatively charged PS particles by amino-groups. Further addition of TEOS will lead to uniform silica growth in all direction, resulting in the formation of silica shell with smooth surface.

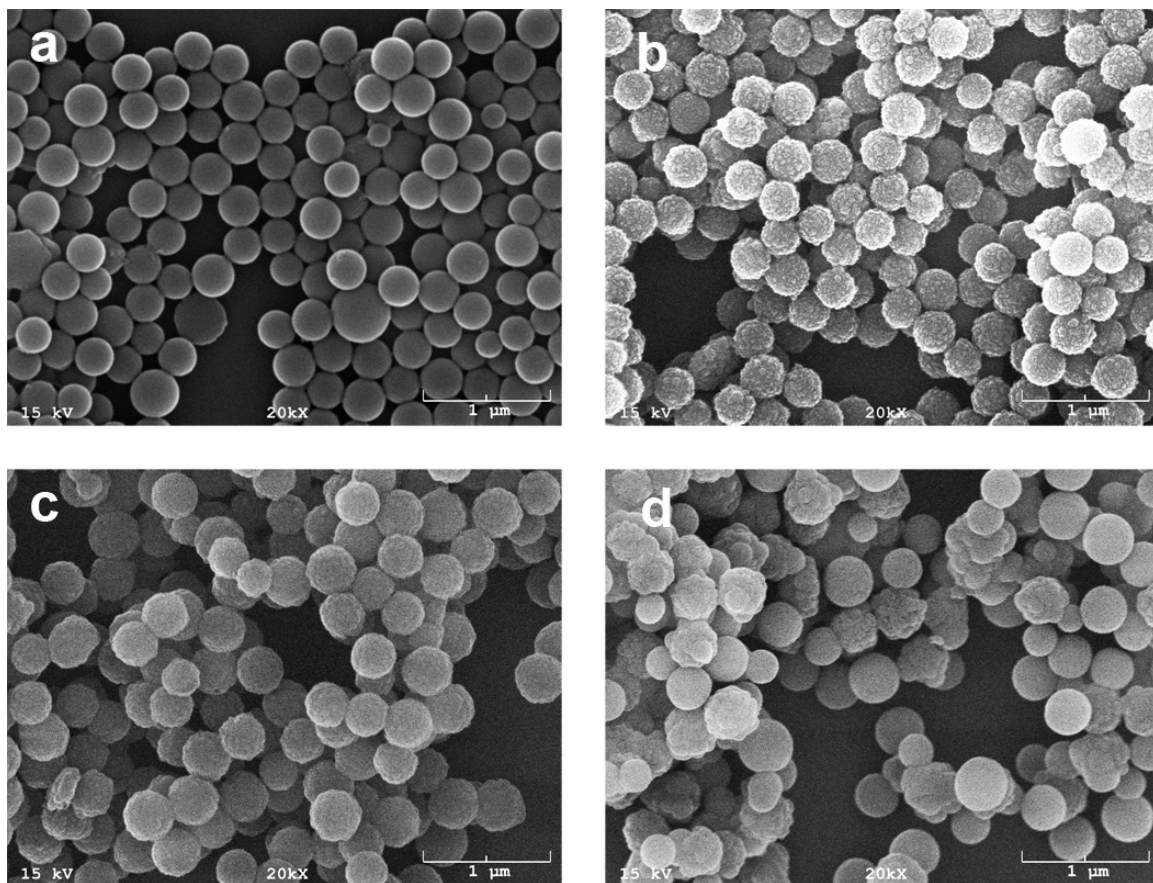


Fig. S1 Low magnification SEM images of hollow silica spheres obtained at various amounts of TEOS. (a) 0.5  $\mu\text{l}$ . (b) 1  $\mu\text{l}$ . (c) 2  $\mu\text{l}$ . (d) 4  $\mu\text{l}$

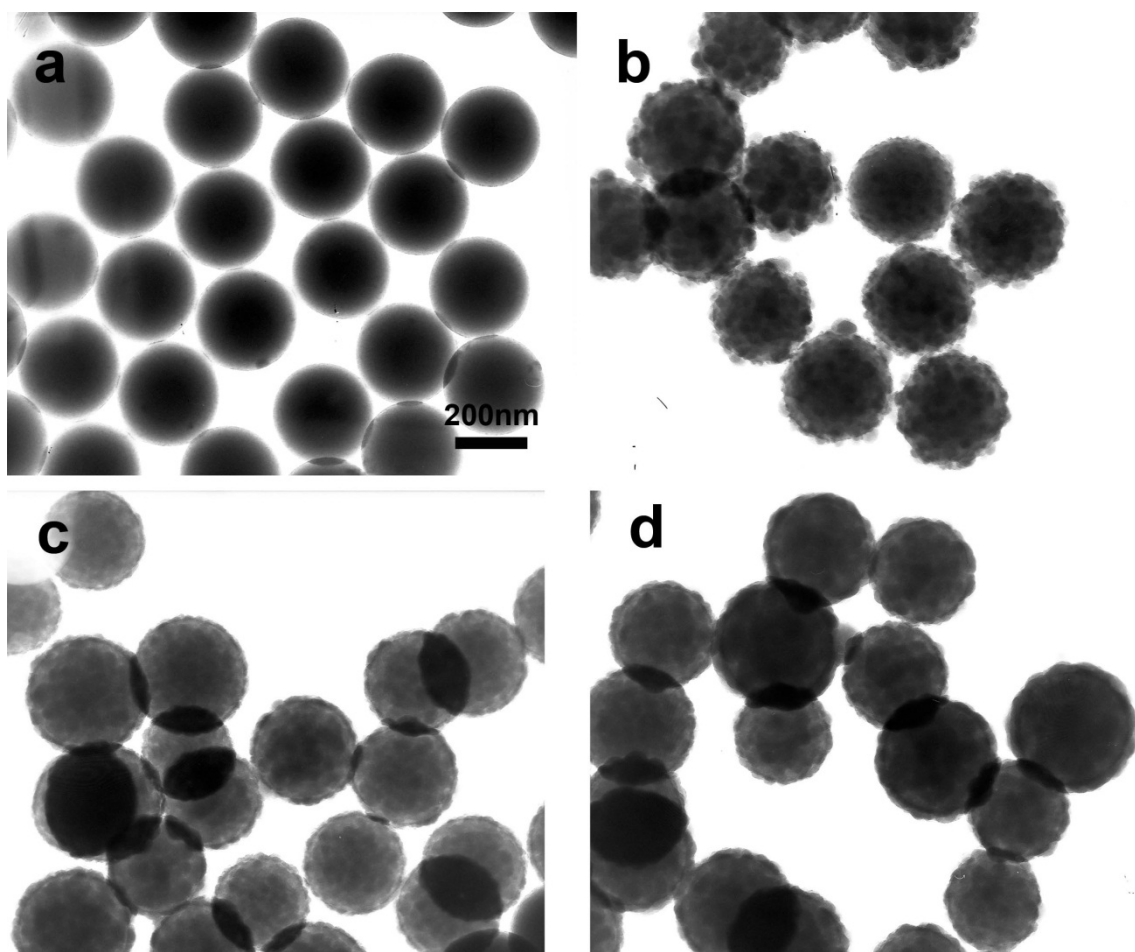


Fig. S2 TEM micrographs of the well-defined silica spheres on amino-PS templates produced with APTES and various amounts of TEOS but no  $\text{CH}_2\text{Cl}_2$ . (a) 0.5  $\mu\text{l}$ . (b) 1  $\mu\text{l}$ . (c) 2  $\mu\text{l}$ . (d) 4  $\mu\text{l}$ .

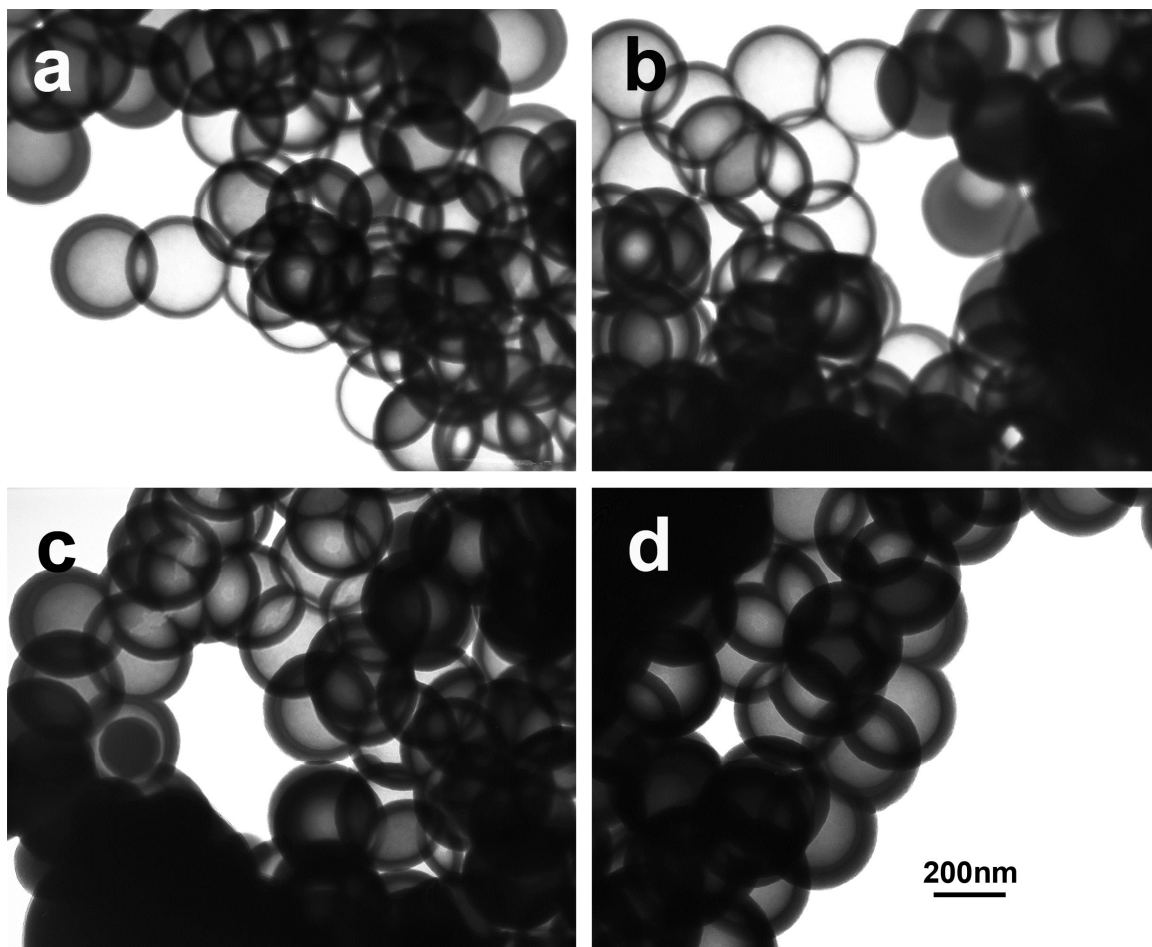


Fig. S3 TEM micrographs of the well-defined hollow silica spheres after core removal with  $\text{CH}_2\text{Cl}_2$  produced using carboxyl-PS spheres as templates and with APTES and various amounts of TEOS. (a) 0.5  $\mu\text{l}$ . (b) 1  $\mu\text{l}$ . (c) 2  $\mu\text{l}$ . (d) 4  $\mu\text{l}$ . A thicker silica wall was observed with a higher concentration of TEOS and the hollow silica spheres kept smooth surfaces.

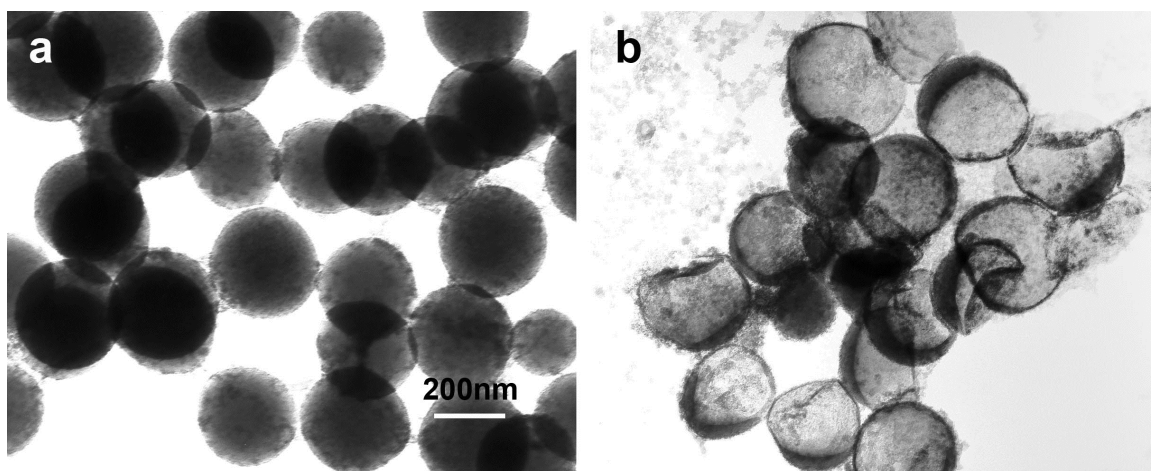


Fig. S4 TEM micrographs of (a) amino-PS/silica spheres mineralized by TEOS only and (b) hollow silica spheres mineralized by TEOS after the addition of  $\text{CH}_2\text{Cl}_2$  to remove PS cores.

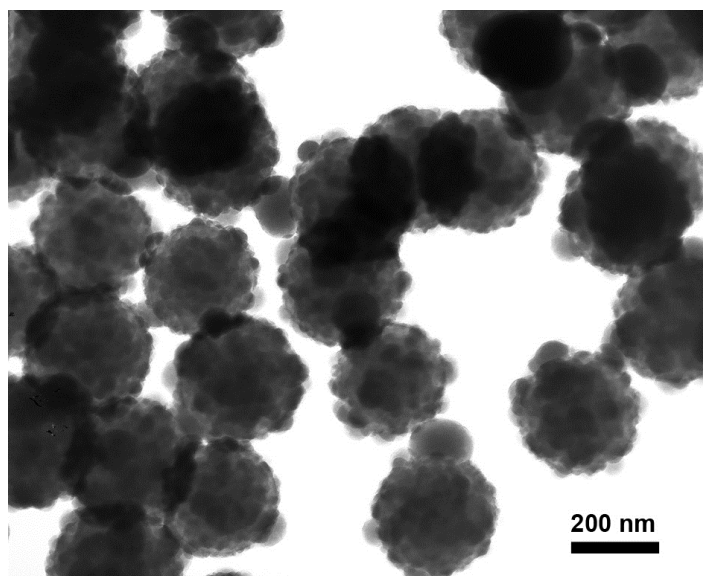


Fig. S5 TEM micrographs of the silica spheres on amino-PS templates. To produce them,  $10^{-2}$  mmol APTES was mixed first with 500  $\mu$ l of PS particles first and then with 1  $\mu$ l of TEOS. A rougher surface of silica spheres with some free silica particles attached was observed.