Supporting Information

Uniform Double-Shell Hollow Microspheres from New Polymer-Backbone-Transition Method as Effective Acoustic Echo Imaging Contrast Agents

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**Figure S1.** The curves of dispersion stability of polymer microbubbles: (a) the stabilization time of the microbubbles with different sizes and the same shell thickness of approximate 65 nm in water as a function of the size; (b) the stabilization time of the microbubbles with the same size of 1.1 μm and different shell thicknesses as a function of shell thickness.

**Figure S2.** TEM images of the organosilica/PNIPAM core/shell microspheres: (a) NIPAM/VOMs = 5:1, 25 mg AIBN, (b) NIPAM/VOMs = 10:1, (c) NIPAM/VOMs = 10:1, 100 mg AIBN, (d) NIPAM/VOMs = 10:1, 200 mg AIBN. All scale bars are 1.0 μm.
**Figure S3** Results from DLS measurements at various temperatures of ds-HPMs with different shell thickness: (a) ds-HPMs-1, (b) ds-HPMs-2, (c) ds-HPMs-3, (d) ds-HPMs-4.

**Figure S4** In vitro ultrasound images of ds-HPMs-2 under B-mode in physiological saline solution with different concentration from 0.0-1.0 mg·mL$^{-1}$ at 4.0 M Hz