## **Supplentary Information**

## Unexpected Behavior of Polydimethylsiloxane / Poly(2-(dimethylamino)ethyl acrylate) (Charged) Amphiphilic Block Copolymers in Aqueous Solution

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**Figure S1.** GPC (THF) chromatograms of PDMS-OH (solid line) and PDMS-PBTC Macro-CTA (dashed line). The peak intensity was normalized to one for clarity.



**Time (h) Figure S2.** Plot of Ln  $[M]_0$  / $[M]_n$  versus time for PDMS-PBTC Macro-CTA mediated polymerization of DMAEA in toluene at 70 °C.  $[DMAEA]_0$  / $[PDMS-PBTC]_0$  / $[AIBN]_0$  = 100:1:0.2, m(DMAEA): m(toluene)= 4:1



**Figure S3.** <sup>1</sup>H NMR spectrum of PDMS<sub>60</sub>-*b*-PDMAEA<sub>60</sub> in CDCl<sub>3</sub> after dialysis against methanol.



**Figure S4.** <sup>1</sup>H NMR spectrum of PDMS<sub>60</sub>-*b*-PDMAEA<sub>60</sub> in *d*-acetone after being dissolved in  $D_2O$  for 240 h.



**Figure S5.** <sup>1</sup>H NMR spectrum of PDMS<sub>60</sub>-*b*-(PDMAEA<sub>25</sub>-*co*-PAA<sub>35</sub>) in D<sub>2</sub>O after dialysis against MilliQ H<sub>2</sub>O.



Figure S6. <sup>1</sup>H NMR spectrum of PDMS<sub>60</sub>-*b*-PTMAIEA<sub>60</sub> in *d*-DMSO after dialysis against H<sub>2</sub>O.



**Figure S7.** FT-IR Spectra of PDMS<sub>60</sub>-*b*-PDMAEA<sub>60</sub>, PDMS<sub>60</sub>-*b*-(PDMAEA<sub>25</sub>-*co*-PAA<sub>35</sub>) and PDMS<sub>60</sub>-*b*-PTMAIEA<sub>60</sub>