Electronic Supplementary Information to the article

## Smart Hydrogels Based on Responsive Star-Block Copolymers

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Figure S1. <sup>1</sup>H-NMR spectra of a)  $(DMA_{150}DEGMA_{100})_4$  in CDCl<sub>3</sub> and b) quaternized  $(qDMA_{150}DEGMA_{100})_4$  in D<sub>2</sub>O.



**Figure S2**. Micro differential scanning calorimetry measurements for a)  $(DMA_{130}DEGMA_{140})_6$  and b)  $(DMA_{130}DEGMA_{60})_6$  at pH 9 (solid lines) compared to  $(DMA_{130})_6$  (dashed line). Measurements were performed at 20 wt% for the diblock copolymer stars and 10 wt% for the homopolymer star, respectively, at a heating rate of 0.5 K/min.



**Figure S3**. Temperature-dependent apparent hydrodynamic radii and polydispersity indices derived from cumulant analysis for  $(DMA_{150}DEGMA_{40})_4$  at a) pH 7 and b) pH 8,  $(DMA_{150}DEGMA_{100})_4$  at c) pH 7 and d) pH 8, and  $(DMA_{130}DEGMA_{60})_6$  at e) pH 7 and f) pH 8. Measurements were performed in buffer solution at 1 g/L and  $\theta = 90^\circ$ .



**Figure S4**. Temperature-dependent apparent hydrodynamic radii and polydispersity indices derived from cumulant analysis for  $(qDMA_{150}DEGMA_{40})_4$  at a) pH 7 and b) pH 8,  $(qDMA_{150}DEGMA_{100})_4$  at c) pH 7 and d) pH 8, and  $(qDMA_{130}DEGMA_{60})_6$  at e) pH 7 and f) pH 8. Measurements were performed in buffer solution at 1 g/L and  $\theta = 90^\circ$ .



**Figure S5**. Temperature-dependent G' and G'' of  $(DMA_{150}DEGMA_{100})_4$  in a) a 15 wt% solution at pH 8.2 and b) a 20 wt% solution at pH 8.4, and of  $(DMA_{130}DEGMA_{60})_6$  in c) a 15 wt% solution at pH 8.2 and d) a 20 wt% solution at pH 8.3.



**Figure S6**. Temperature-dependent *G'* and *G''* of  $(DMA_{150}DEGMA_{100})_4$  in a) a 15 wt% solution at pH 8.9 and b) a 20 wt% solution at pH 8.9, and of  $(DMA_{130}DEGMA_{60})_6$  in c) a 15 wt% solution at pH 9.0 and d) a 20 wt% solution at pH 8.7.



**Figure S7**. Temperature-dependent G' and G'' of  $(qDMA_{150}DEGMA_{100})_4$  in a) a 5 wt% solution and b) a 10 wt% solution, and of  $(qDMA_{130}DEGMA_{60})_6$  in c) a 5 wt% solution and d) a 10 wt% solution.