## **Supplementary Information**

## **Rational Design of Hydrogen Bonds for Driving Thermo-responsive Phase**

## Transition and Assembly Behavior of Block Copolymer in Water

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Scheme S1 Schematic illustration of the preparation of PEG-b-PAAm-b-PAAc block copolymer.



Scheme S2 Schematic illustration of the preparation of PEG-b-P(AAm-co-AAc) random copolymer.



**Fig. S1** <sup>1</sup>H NMR spectra of PEG<sub>45</sub>-*b*-P(AAm<sub>176</sub>-*co*-AAc<sub>67</sub>) random copolymer and PEG<sub>45</sub>-*b*-PAAm<sub>174</sub>-*b*-PAAc<sub>70</sub> block copolymer.



Fig. S2 (a) FT-IR spectra of BSPA; (b) <sup>1</sup>H NMR spectra of BSPA.



Fig. S3 Size exclusion chromatography (SEC) spectra of PEG<sub>45</sub>-b-PAAm<sub>178</sub>-b-PAAc<sub>134</sub>.



**Fig. S4** (a) FTIR spectra of PEG-*b*-PAAc, PEG-*b*-P(AAc-*co*-GMA), PEG-*b*-PAAm-*b*-P(AAc-*co*-GMA); (b) FTIR spectra of PEG-*b*-PAAm-*b*-P(AAc-*co*-GMA) with different feed ratios of AAc and AAm (1:1, 1:2 and 1:3); (c) <sup>1</sup>H NMR spectra of PEG<sub>45</sub>-PAAc<sub>134</sub>, PEG<sub>45</sub>-*b*-P(AAc<sub>140</sub>-*co*-GMA<sub>21</sub>); (d) <sup>1</sup>H NMR spectra of PEG-*b*-PAAm-*b*-P(AAc-*co*-GMA) with different feed ratios of AAc and AAm (1:1, 1:2 and 1:3).



Fig. S5 Temperature-dependent transmittance of 5 mg/ml  $PEG_{45}$ -b-PAAm<sub>178</sub>-b-PAAc<sub>134</sub> aqueous solutions at 500 nm at different pH values.



**Fig. S6** Salt-dependent UCST behaviour of PEG<sub>45</sub>-*b*-PAAm<sub>176</sub>-*b*-P(AAc<sub>140</sub>-*co*-GMA<sub>21</sub>) block copolymer measured at 500 nm.