## **Electronic Supporting Information**

## Hydrogels from Amphiphilic Star Block Copolypeptides

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Figure S1. <sup>1</sup> H-NMR spectrum of 8-PZLL<sub>40</sub> (CDCl<sub>3</sub>).



Figure S2. <sup>1</sup> H-NMR spectrum of 8-PZLL<sub>40</sub>-*b*-PBLT<sub>5</sub> (CDCl<sub>3</sub>).



Figure S3. <sup>1</sup> H-NMR spectrum of 8-PLL<sub>40</sub>-*b*-PLT<sub>5</sub> (D<sub>2</sub>O).



Figure S4: SEC trace of; 8-PZLL<sub>40</sub>-b-PLA<sub>5</sub> (A) and 8-PZLL<sub>40</sub> (B).



Figure S5: SEC trace of; 8-PZLL<sub>40</sub>-b-PLT<sub>5</sub> (A) and 8-PZLL<sub>40</sub> (B).



Figure S6: SEC trace of; 1-PZLL<sub>148</sub>-b-PBLT<sub>30</sub> (A) and 1-PZLL<sub>148</sub> (B).



Figure S7: FT-IR spectra with assigned amide bands.



Figure S8: Swelling ratio of star diblock copolypeptides (3.0 wt.%) as a function of increasing ionic concentration.



Figure S9: Strain based recovery of gel strength [sheared at  $\gamma = 1$ ,  $\omega = 6$  rad s<sup>-1</sup> for 8 mins before switching shear to  $\gamma = 0.1$ ,  $\omega = 6$  rad s<sup>-1</sup> for 24 mins]. 8-PLL<sub>40</sub>-*b*-PLPA<sub>5</sub> ( $\bullet$ ), 8-PLL<sub>40</sub>-*b*-PLV<sub>5</sub> ( $\bullet$ ), 8-PLL<sub>40</sub>-*b*-PLT<sub>5</sub> ( $\bullet$ ).



Figure S10: Image of 8-PLL<sub>40</sub>-b-PLPA<sub>5</sub> hydrogel loaded in a syringe.



Figure S11: Image of 8-PLL<sub>40</sub>-b-PLPA<sub>5</sub> with in a syringe with a 21G needle attached.



Figure S12: Circular dichroism (CD) spectrum of 8-arm PLL in water.