Supporting Information for:

Crucial Impact of Residue Chirality on the Gelation Process and Biodegradability of Thermoresponsive Polypeptide Hydrogels

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Figure S1. ¹H-NMR spectra of (A) ELG and (B) EDG in D₂O, as well as (C) ELG NCA and (D) EDG NCA in CDCl₃.



Figure S2. FT-IR spectra of (A) EG₄₅LG_{16.9}, (B) EG₄₅(LG_{0.75}DG_{0.25})_{15.6}, (C) EG₄₅(LG_{0.5}DG_{0.5})_{15.4},

(D) $EG_{45}(LG_{0.25}DG_{0.75})_{16.6}$, and (E) $EG_{45}DG_{16.0}$.



Figure S3. GPC traces of (A) EG₄₅LG_{16.9}, (B) EG₄₅(LG_{0.75}DG_{0.25})_{15.6}, (C) EG₄₅(LG_{0.5}DG_{0.5})_{15.4}, (D) EG₄₅(LG_{0.25}DG_{0.75})_{16.6}, and (E) EG₄₅DG_{16.0}.



Figure S4. Temperature dependent storage moduli (G') and loss moduli (G'') of (A) $EG_{45}LG_{16.9}$ hydrogel, (B) $EG_{45}(LG_{0.75}DG_{0.25})_{15.6}$ hydrogel, (C) $EG_{45}(LG_{0.5}DG_{0.5})_{15.4}$ hydrogel, (D) $EG_{45}(LG_{0.25}DG_{0.75})_{16.6}$ hydrogel, and (E) $EG_{45}DG_{16.0}$ hydrogel.



Figure S5. Typical excitation spectra of pyrene in copolymer aqueous solutions at different concentrations (λ_{em} =390 nm; A: EG₄₅LG_{16.9}; B: EG₄₅(LG_{0.75}DG_{0.25})_{15.6}; C: EG₄₅(LG_{0.5}DG_{0.5})_{15.4}; D: EG₄₅(LG_{0.25}DG_{0.75})_{16.6}; E: EG₄₅DG_{16.0}). The unit Mcps represents million counts per second.



Figure S6. ¹³C-NMR spectra of copolymers in D₂O at diverse temperatures. (A) $EG_{45}LG_{16.9}$, 6.0 wt%; (B) $EG_{45}(LG_{0.75}DG_{0.25})_{15.6}$, 4.0 wt%; (C) $EG_{45}(LG_{0.5}DG_{0.5})_{15.4}$, 4.0 wt%; (D) $EG_{45}(LG_{0.25}DG_{0.75})_{16.6}$, 4.0 wt%; (E) $EG_{45}DG_{16.0}$, 6.0 wt%.



Figure S7. In vitro cytocompatibility L929 cells measured with CCK-8 after cultured with five copolymer solutions (n=5; A: EG₄₅LG_{16.9}; B: EG₄₅(LG_{0.75}DG_{0.25})_{15.6}; C: EG₄₅(LG_{0.5}DG_{0.5})_{15.4}; D: EG₄₅(LG_{0.25}DG_{0.75})_{16.6}; E: EG₄₅DG_{16.0}).



Figure S8. In vitro degradation profiles of hydrogels (6.0 wt%) incubated in PBS (A) or PBS with proteinase K (5 U/mL) (B).



Figure S9. Photographs of the in vivo degradation of $EG_{45}LG_{16.9}$ hydrogels (6 wt%) after various time intervals. Triplicate tests were performed at each time point.



Figure S10. Photographs of the in vivo degradation of $EG_{45}(LG_{0.75}DG_{0.25})_{15.6}$ hydrogels (6 wt%) after various time intervals.



Figure S11. Photographs of the in vivo degradation of $EG_{45}(LG_{0.5}DG_{0.5})_{15.4}$ hydrogels (6 wt%) after various time intervals.



Figure S12. Photographs of the in vivo degradation of $EG_{45}(LG_{0.25}DG_{0.75})_{16.6}$ hydrogels (6 wt%) after various time intervals.



Figure S13. Photographs of the in vivo degradation of EG₄₅DG_{16.0} hydrogels (6 wt%) after various time intervals.



Figure S14. Images of H&E-stained sections of tissue surrounding the injected $EG_{45}LG_{16.9}$ hydrogels at different time points. G: gel, T: tissue.



Figure S15. Images of H&E-stained sections of tissue surrounding the injected $EG_{45}(LG_{0.75}DG_{0.25})_{15.6}$ hydrogels at different time points. G: gel, T: tissue.



Figure S16. Images of H&E-stained sections of tissue surrounding the injected $EG_{45}(LG_{0.5}DG_{0.5})_{15.4}$ hydrogels at different time points. G: gel, T: tissue.



Figure S17. Images of H&E-stained sections of tissue surrounding the injected $EG_{45}(LG_{0.25}DG_{0.75})_{16.6}$ hydrogels at different time points. G: gel, T: tissue.



Figure S18. Images of H&E-stained sections of tissue surrounding the injected $EG_{45}DG_{16.0}$ hydrogels at different time points. G: gel, T: tissue.