Supporting Information for Manuscript Entitled with

Hydrogels assembled from star-shaped polypeptides with

dendrimer as core

Yong Shen^a, Shusheng Zhang^a, Yaoming Wan^a, Wenxin Fu^a and Zhibo Li^{a,b,*}

^a Beijing National Laboratory for Molecular Sciences (BNLMS), Laboratory of Polymer Physics and Chemistry, Institute of Chemistry, Chinese Academy of Sciences, Beijing 100190, China.

^b School of Polymer Science and Engineering, Qingdao University of Science and Technology, Qingdao 266042, China.

E-mail: zbli@iccas.ac.cn



Scheme S1. Chemical structure of G2-[NH₂]₈.



Scheme S2. Chemical structure of G4-[NH₂]₃₂.



Figure S1. (a) ¹H and (b) ¹³C NMR spectra of $G2-[NH_2]_8-(EG_2Glu)_{15}$.



Figure S2. (a) Strain sweeps for the G2-[NH₂]₈-(EG₂Glu)₃₀ aqueous solution at angular frequency $\omega = 1$ rad/s; (b) Storage modulus G' (solid symbols) and loss modulus G'' (open symbols) as a function of angular frequency for the same samples ($\gamma = 0.01$) at two different concentrations: (\blacktriangle) 3.0 wt %, (\bullet) 4.0 wt %.



Figure S3. (a) Strain sweeps for the G4-[NH₂]₃₂-(EG₂Glu)₁₅ aqueous solution at angular frequency $\omega = 1$ rad/s; (b) Storage modulus G' (solid symbols) and loss modulus G'' (open symbols) as a function of angular frequency for the same samples ($\gamma = 0.01$) at three different concentrations: (**■**) 2.0 wt %, (**▲**) 3.0 wt %, (**●**) 4.0 wt %.



Figure S4. (a) Strain sweeps for the G4-[NH₂]₃₂-(EG₂Glu)₃₀ aqueous solution at angular frequency $\omega = 1$ rad/s; (b) Storage modulus G' (solid symbols) and loss modulus G'' (open symbols) as a function of angular frequency for the same samples ($\gamma = 0.01$) at three different concentrations: (**•**) 2.0 wt %, (**•**) 3.0 wt %, (**•**) 4.0 wt %.



Figure S5. Comparison of (a) CD and (b) FT-IR spectra of $G2-[NH_2]_8-(EG_2Glu)_n$ and $G4-[NH_2]_{32}-(EG_2Glu)_n$ (n = 15, 30).



Figure S6. Deconvolution of amide I band in FT-IR spectra of $G2-[NH_2]_8-(EG_2Glu)_{15}$ (a), G2-[NH₂]₈-(EG₂Glu)₂₀ (b), G2-[NH₂]₈-(EG₂Glu)₃₀ (c), G2-[NH₂]₈-(EG₂Glu)₅₀ (d), G4-[NH₂]₃₂-(EG₂Glu)₁₅ (e), and G4-[NH₂]₃₂-(EG₂Glu)₃₀ (f).



Figure S7. TEM images of G2- $[NH_2]_8$ - $(EG_2Glu)_{30}$ aqueous solutions: (**a**) 0.03 wt%, (**b**) 0.3 wt%, (**c**) 3 wt%.

Table S1. The secondary structure contents calculated by deconvolution of amide I band in FT-IR spectra.

Sample	α-helix	β-sheet	Antiparallel β-sheet	Random coil
G2-[NH ₂] ₈ -(EG ₂ Glu) ₁₅	54.6%	21.7%	6.2%	17.5%
$G2-[NH_2]_8-(EG_2Glu)_{20}$	57.9%	18.8%	5.8%	17.5%
G2-[NH ₂] ₈ -(EG ₂ Glu) ₃₀	62.4%	14.9%	5.1%	17.6%
G2-[NH ₂] ₈ -(EG ₂ Glu) ₅₀	60.6%	15.9%	5.3%	18.2%
G4-[NH ₂] ₃₂ -(EG ₂ Glu) ₁₅	52.1%	23.7%	6.7%	17.4%
G4-[NH ₂] ₃₂ -(EG ₂ Glu) ₃₀	60.1%	17.0%	5.5%	17.4%