## **Supplementary Materials**

## Effects of Hydrophilic-Lipophilic Balance of Alternating Peptides on Self-Assembly and Thermo-Responsive Behaviors

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Figure S1: DOSY correlations of Poly(Gly-*alter*-L-Val) (400 MHz, TFA-*d*, 298 K) and the attenuation curve.



**Figure S2:** DOSY correlations of **Poly(Gly-***alter***-L-Val)** (400 MHz, D<sub>2</sub>O, 298 K) and the attenuation curve.



Figure S3: IR spectrum of Poly(Gly-alter-L-Val) (ATR).





Figure S5: <sup>13</sup>C NMR spectrum of Et-Pep (100 MHz, CDCl<sub>3</sub>, 298 K).



Figure S6: DOSY correlations of Et-Pep (400 MHz, TFA-d, 298 K) and the attenuation curve.



Figure S7: DOSY correlations of Et-Pep (400 MHz,  $D_2O$ , 298 K) and the attenuation curve.



Figure S8: IR spectrum of Et-Pep (ATR).



Figure S9: <sup>1</sup>H NMR spectrum of HE-Pep (400 MHz, CDCl<sub>3</sub>, 298 K).





Figure S11: DOSY correlations of HE-Pep (400 MHz, TFA-d, 298 K) and the attenuation curve.



Figure S12: DOSY correlations of HE-Pep (400 MHz, D<sub>2</sub>O, 298 K) and the attenuation curve.



Figure S13: IR spectrum of HE-Pep (ATR).



Figure S15: <sup>13</sup>C NMR spectrum of Pr-Pep (100 MHz, CDCl<sub>3</sub>, 298 K).



Figure S16: DOSY correlations of Pr-Pep (400 MHz, TFA-*d*, 298 K) and the attenuation curve.



Figure S17: DOSY correlations of Pr-Pep (400 MHz, D<sub>2</sub>O, 298 K) and the attenuation curve.



Figure S18: IR spectrum of Pr-Pep (ATR).





Figure S20: <sup>13</sup>C NMR spectrum of Me-Pep (100 MHz, CDCl<sub>3</sub>, 298 K).



Figure S21: DOSY correlations of Me-Pep (400 MHz, TFA-d, 298 K) and the attenuation curve.



Figure S22: DOSY correlations of Me-Pep (400 MHz, D<sub>2</sub>O, 298 K) and the attenuation curve.



Figure S23: IR spectrum of Me-Pep (ATR).



Figure S24. UV-vis spectra of Poly(Gly-alter-L-Val) in H<sub>2</sub>O at various concentrations at 25 °C.



Figure S25. UV-vis spectra of Et-Pep in H<sub>2</sub>O at various concentrations at 25 °C.



Figure S26. UV-vis spectra of Et-Pep in H<sub>2</sub>O at various temperatures (1.5 wt%).

Me-Pep."						
Alternating Peptide	CMC	Cloud point	Rh in D2O	Rh in TFA-d	R <sub>water</sub> / Rtfa	
	(Wt 70)	(°C)	(R <sub>water</sub> , nm)	(Rtfa, nm)		
Poly(Gly- <i>alter</i> -L-Val)	0.2	15	0.600 ± 0.031	0.907 ± 0.015	0.66	
Et-Pep	0.2	10	0.527 ± 0.005	0.556 ± 0.012	0.95	
MePep	0.2	15	0.538 ± 0.015	0.542 ± 0.010	0.99	

**Table S1.** CMC, cloud point, and hydrodynamic radius (*R*<sub>h</sub>) of **Poly(Gly-***alter***-L-Val)**, **Et-Pep**, and **Me-Pep**.<sup>*a*</sup>

<sup>*a*</sup> Standard deviations are from three samples.