

## **Multi-responsive physical gels formed by a biosynthetic asymmetric triblock protein polymer and a polyanion**

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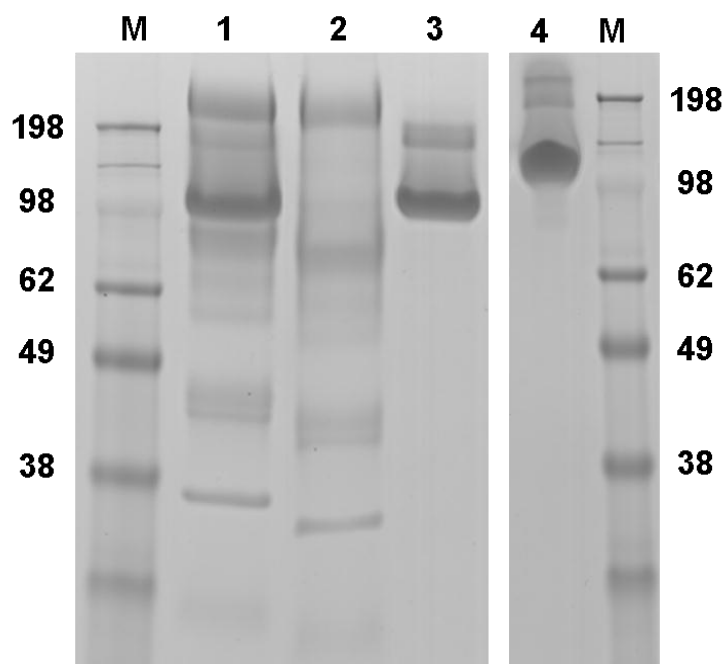
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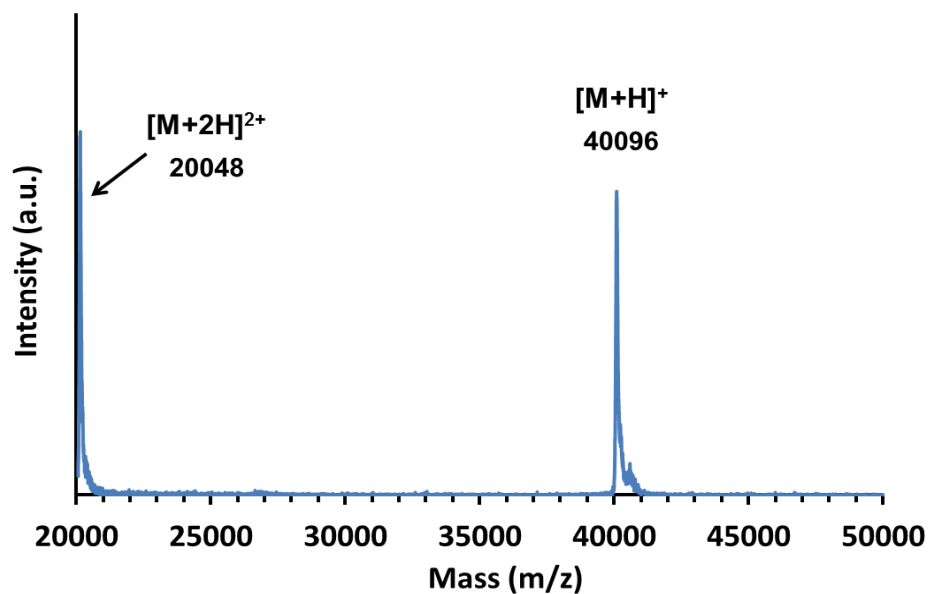
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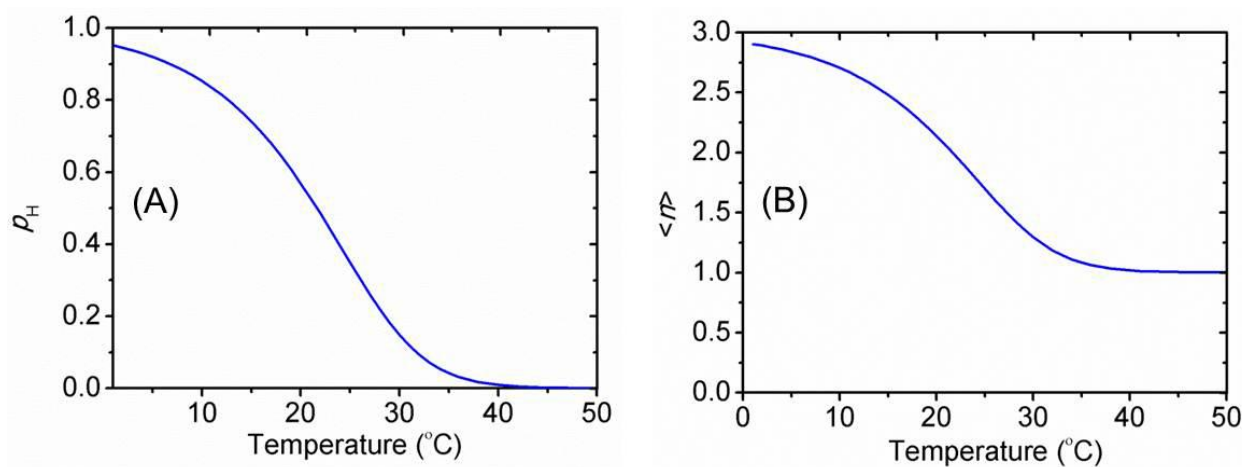
## SUPPORTING INFORMATION



**Figure S1:** SDS-PAGE of TR4H during purification process (M) Protein marker, (lane 1) cell-free fermentation broth, (lane 2) supernatant of 40 % ammonium sulfate saturation 1x, (lane 3 ) protein precipitation with 40 % ammonium sulfate saturation 1x , (lane 4) purified TR4H by precipitation with 40 % ammonium sulfate saturation twice.

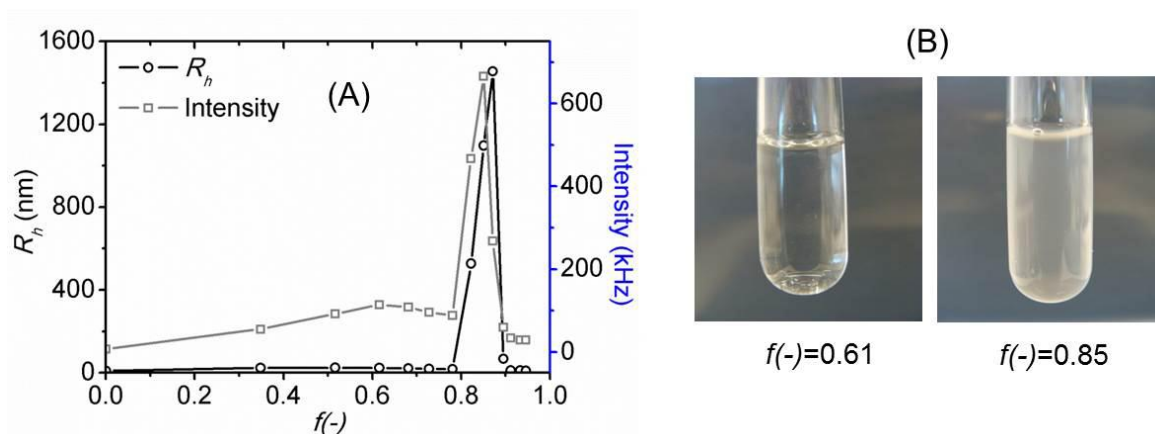


**Figure S2:** MALDI-TOF of purified TR4H. Singly and doubly charged molecular ions are indicated.

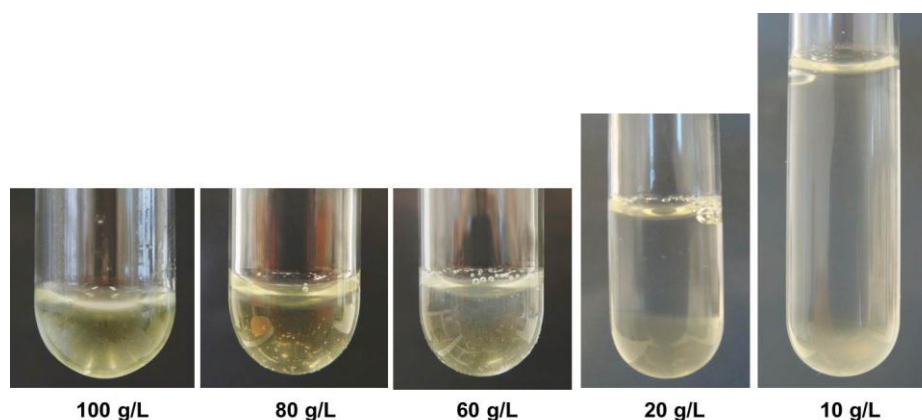


**Figure S3:** Equilibrium calculations of triple helix formation at a concentration  $C$  of 0.125 mM (corresponding to the conditions in Figure 2B). Calculations were done with a temperature-dependent equilibrium constant for helix formation,  $K=[H]/[T]^3$ , with  $[H]$  the concentration of triple helices and  $[T]$  the concentration of free T blocks. The temperature dependence of  $K$  was published previously [Skrzeszewska *et al.*; *Soft Matter* 2009, 5, 2057] and was found to follow van 't Hoff's equation,  $K=K_0 \times \exp(-\Delta H/RT)$  with the enthalpy  $\Delta H=-$

250 kJ/mol and the pre-exponential factor  $K_0=4.1\times 10^{37} \text{ M}^{-2}$ , and with  $R$  the gas constant. (A) The calculated fraction of end blocks in triple helices as a function of temperature; the broad melting curve is in agreement with the light scattering data of Figure 2B. (B) The calculated weight-averaged cluster size,  $\langle n \rangle = ([T] + 9[H])/C$ , as a function of temperature.



**Figure S4:** (A) Light scattering titration of charged TR4H with PSS in 10 mM phosphate buffer pH 3, (B) clear sample at stoichiometric peak ( $f^- = 0.61$ ) and visible large aggregates at the second peak at  $f^- = 0.85$ .



**Figure S5:** Phase separation of temperature-induced charge-driven TR4H/PSS micelle solutions at different protein concentrations.