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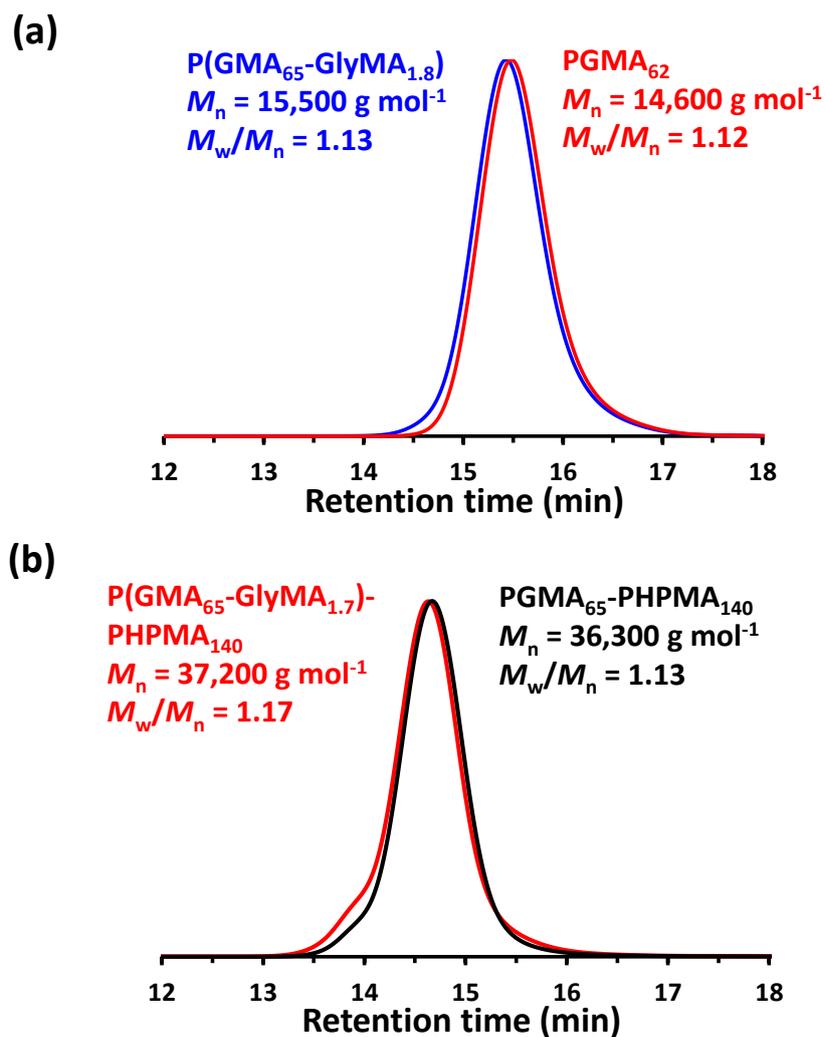
### **Cationic Disulfide-Functionalized Worm Gels**

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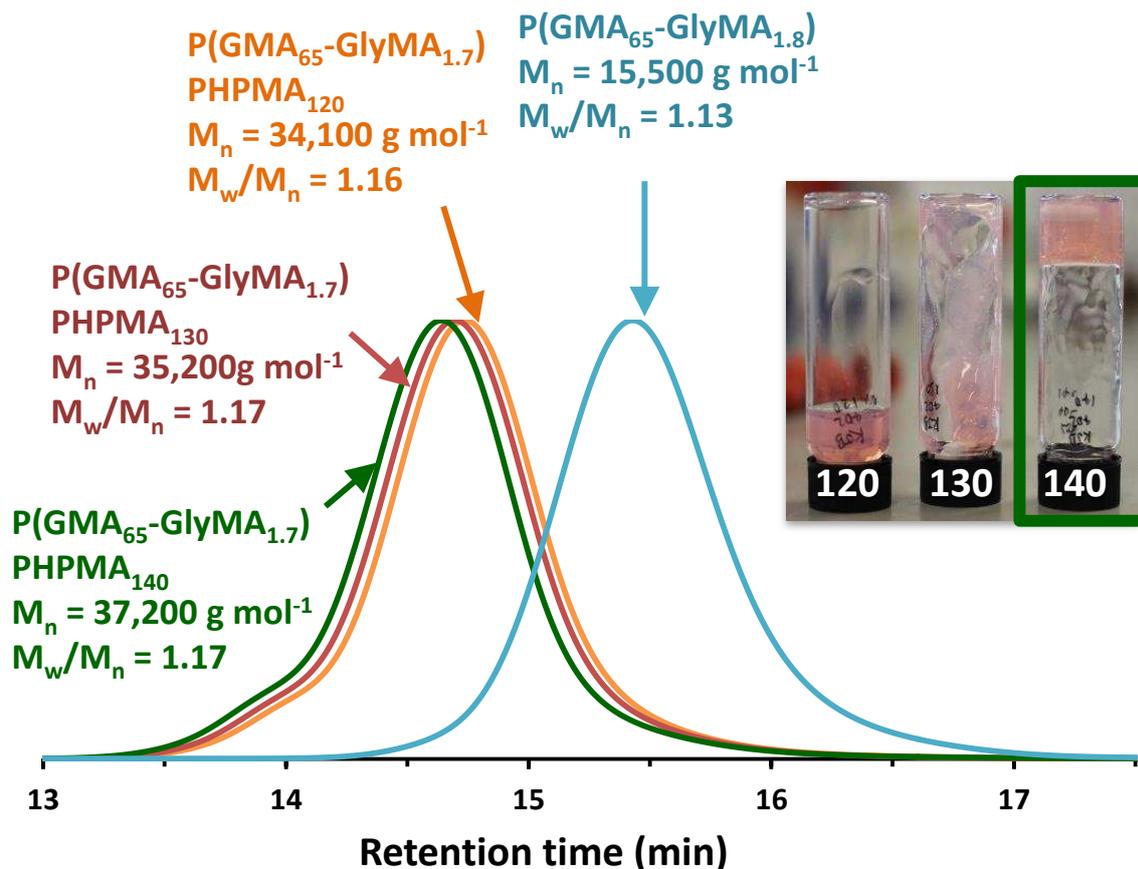
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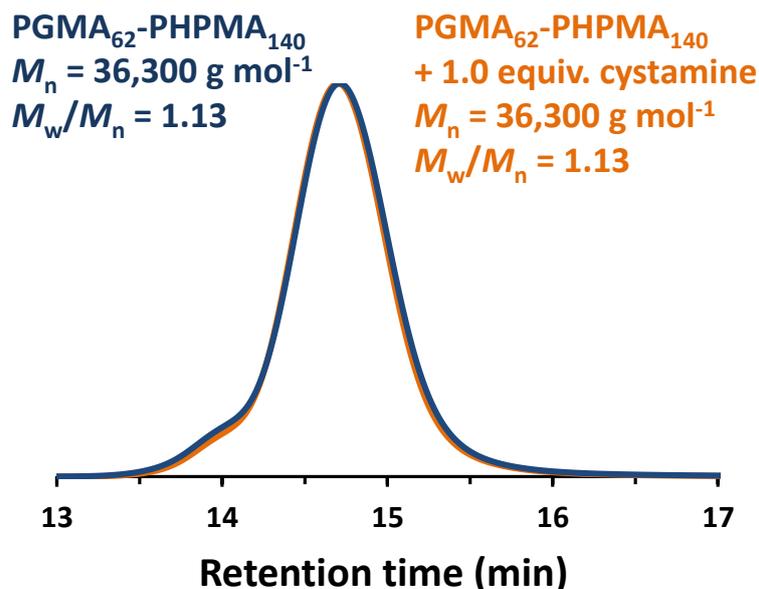
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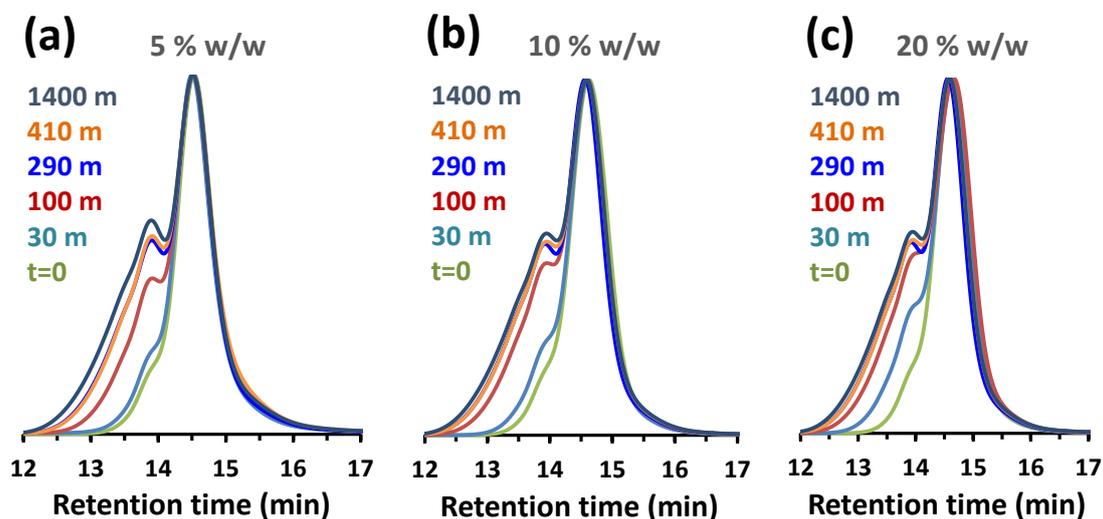
**Figure S1.** (a) DMF GPC chromatograms obtained for P(GMA<sub>65</sub>-stat-GlyMA<sub>1.8</sub>) and PGMA<sub>62</sub> macro-CTAs, both synthesized by RAFT solution (co)polymerization at 55 % w/w and 70 °C in ethanol, using a CTA/initiator molar ratio of 4.0. (b) DMF GPC chromatograms obtained for P(GMA<sub>65</sub>-stat-GlyMA<sub>1.7</sub>)-PHPMA<sub>140</sub> and PGMA<sub>62</sub>-PHPMA<sub>140</sub> worm gels, both synthesized by RAFT aqueous dispersion polymerization of HPMA at 20 % w/w and 50 °C using a CTA/initiator molar ratio of 4.0.



**Figure S2.** DMF GPC chromatograms obtained for P(GMA<sub>65</sub>-stat-GlyMA<sub>1.8</sub>) macro-CTA and for a series of P(GMA<sub>65</sub>-stat-GlyMA<sub>1.7</sub>)-PHPMA<sub>y</sub> diblock copolymers synthesized by RAFT aqueous dispersion polymerization of HPMA at 50 °C in water, at 20 % w/w solids utilizing a macro-CTA/VA-044 molar ratio of 4.0. The digital photographs of these diblock copolymers at 20 % w/w are shown, demonstrating that a free-standing gel was only formed at 22 °C for a diblock copolymer composition of P(GMA<sub>65</sub>-stat-GlyMA<sub>1.7</sub>)-PHPMA<sub>140</sub> (white text corresponds to the PHPMA DP of these copolymers). This suggests the presence of a pure worm phase, as confirmed by the representative TEM image of a 0.2 % w/w dispersion of P(GMA<sub>65</sub>-stat-GlyMA<sub>1.7</sub>)-PHPMA<sub>140</sub>, diluted at 22 °C in water (see Figure 7b in the main text).



**Figure S3.** DMF GPC chromatograms obtained for PGMA<sub>62</sub>-PHPMA<sub>140</sub>, and PGMA<sub>62</sub>-PHPMA<sub>140</sub> + cystamine at 10 % w/w, after stirring for 24 h at 22 °C (equivalent to cystamine/epoxide molar ratio = 1.0 for P(GMA<sub>65</sub>-stat-GlyMA<sub>1.7</sub>)-HPMA<sub>140</sub>), as a control experiment.



**Figure S4.** DMF GPC chromatograms for cystamine functionalization of P(GMA<sub>65</sub>-stat-GlyMA<sub>1.7</sub>)-PHPMA<sub>140</sub> worm gel ([cystamine]/[epoxide] = 0.50) at (a) 5 % w/w, (b) 10 % w/w, and (c) 20 % w/w solids, stirred for 24 h at 22 °C.