

## Supporting Information for:

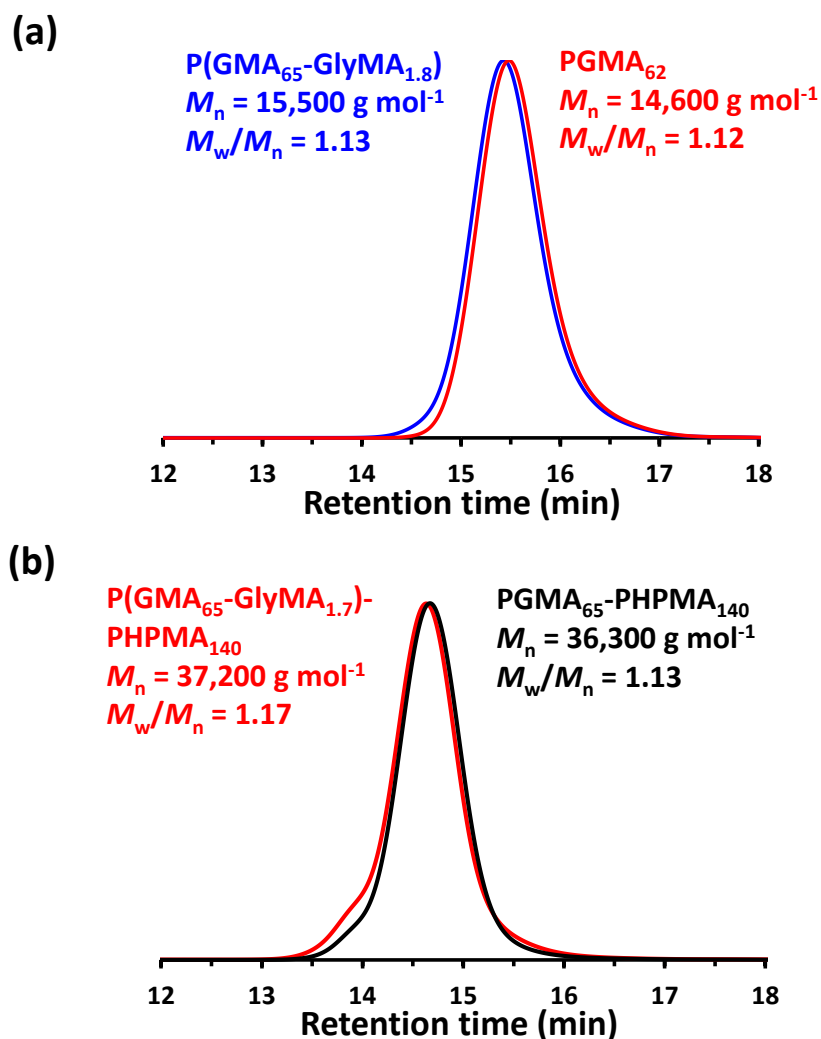
### Cationic Disulfide-Functionalized Worm Gels

*L. P. D. Ratcliffe,<sup>a,\*</sup> K. J. Bentley,<sup>a</sup> R. Wehr,<sup>a</sup> N. J. Warren,<sup>b,\*</sup> B. R. Saunders<sup>c</sup>  
and S. P. Armes<sup>a,\*</sup>*

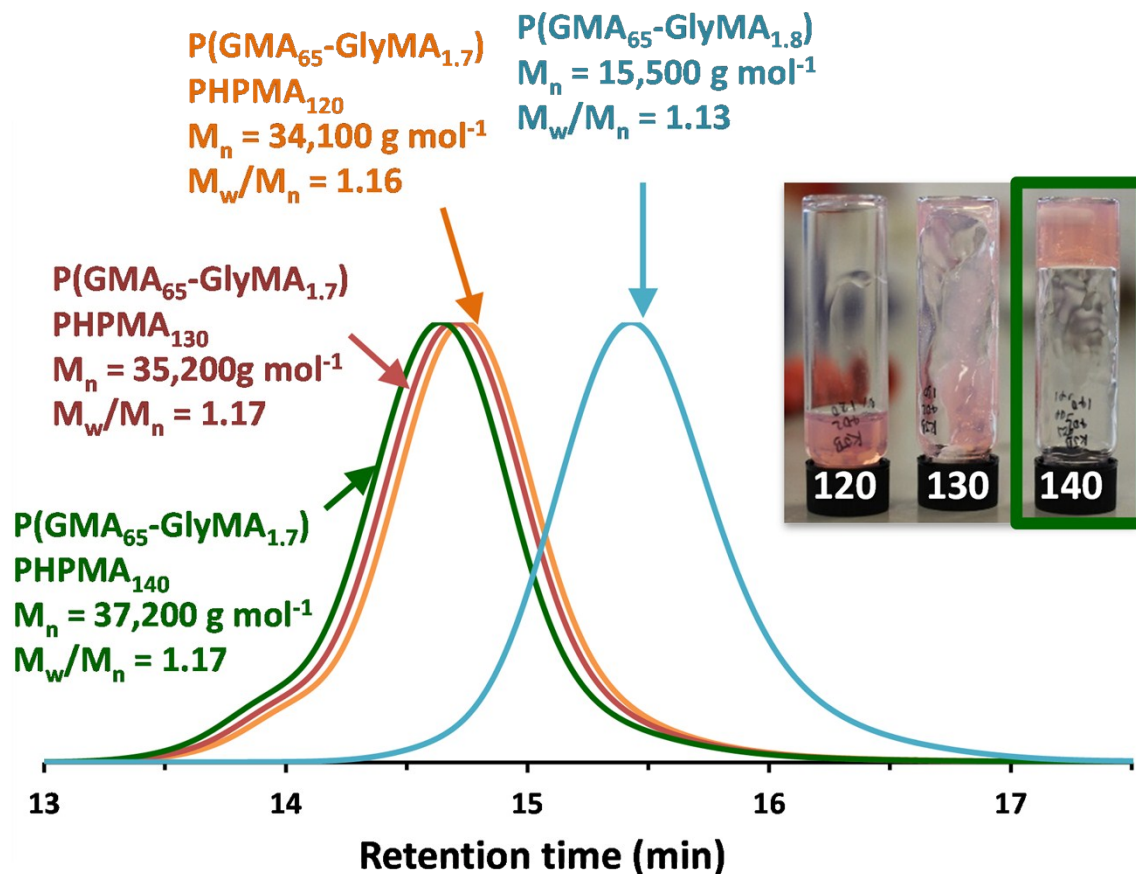
<sup>a</sup>Dainton Building, Department of Chemistry, University of Sheffield,  
Brook Hill, Sheffield, South Yorkshire, S3 7HF, UK

<sup>b</sup>School of Chemical and Process Engineering, University of Leeds, Leeds LS2 9JT

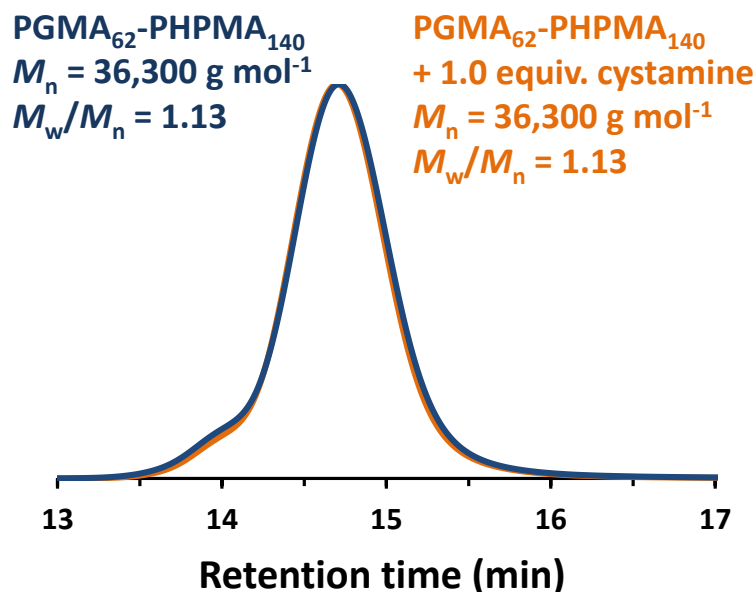
<sup>c</sup>School of Materials, The University of Manchester, MSS Tower, Manchester, M13 9PL, UK



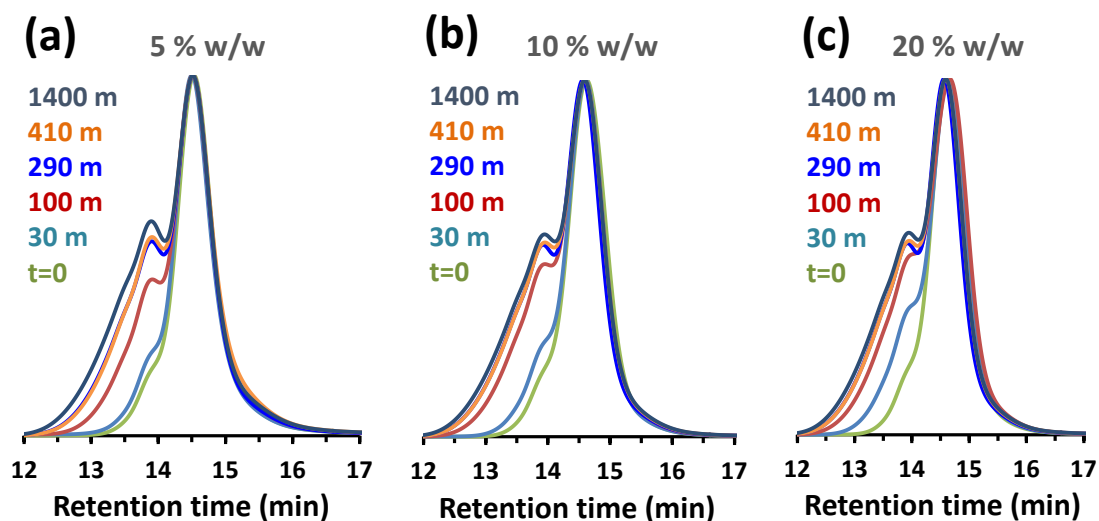
**Figure S1.** (a) DMF GPC chromatograms obtained for P(GMA<sub>65</sub>-*stat*-GlyMA<sub>1.8</sub>) and PGMA<sub>62</sub> macro-CTAs, both synthesized by RAFT solution (co)polymerization at 55 % w/w and 70 °C in ethanol, using a CTA/initiator molar ratio of 4.0. (b) DMF GPC chromatograms obtained for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