

SUPPORTING INFORMATION

**“Off-the-Shelf” Thermoresponsive Hydrogel Design: Tuning Hydrogel Properties  
by Mixing Precursor Polymers with Different Lower-Critical Solution  
Temperatures**

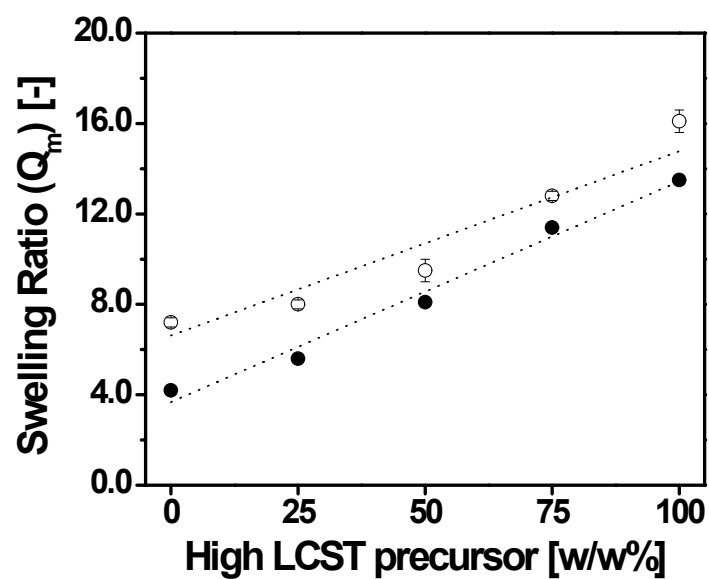
Emilia Bakaic<sup>‡</sup>, Niels M.B. Smeets<sup>‡</sup>, Helen Dorrington, and Todd Hoare\*

<sup>‡</sup>These authors contributed equally.

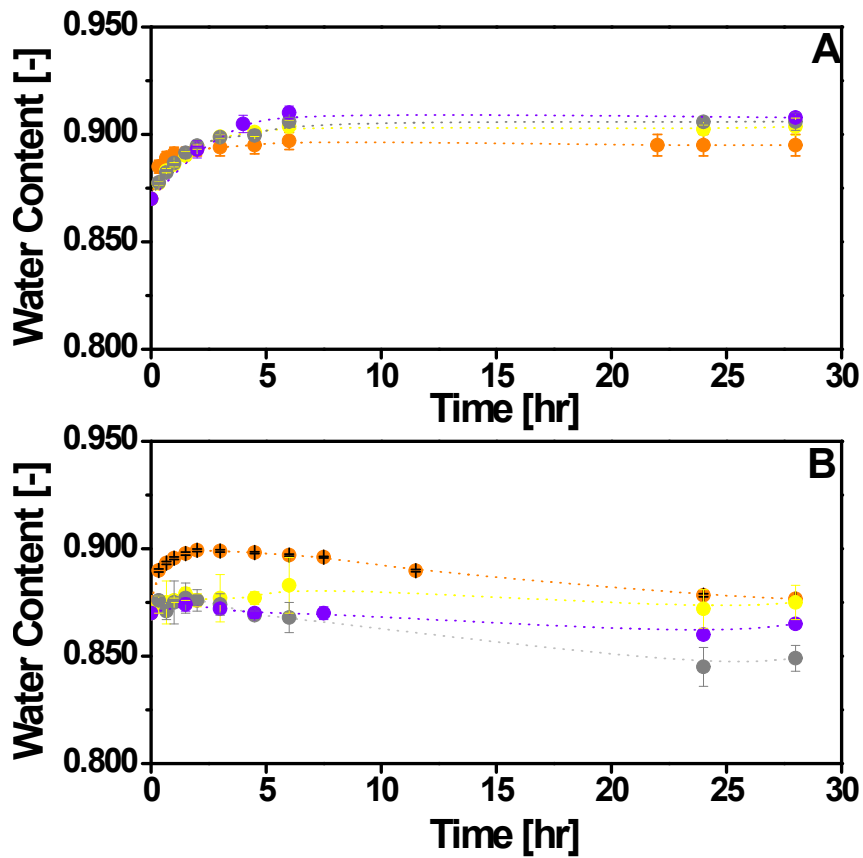
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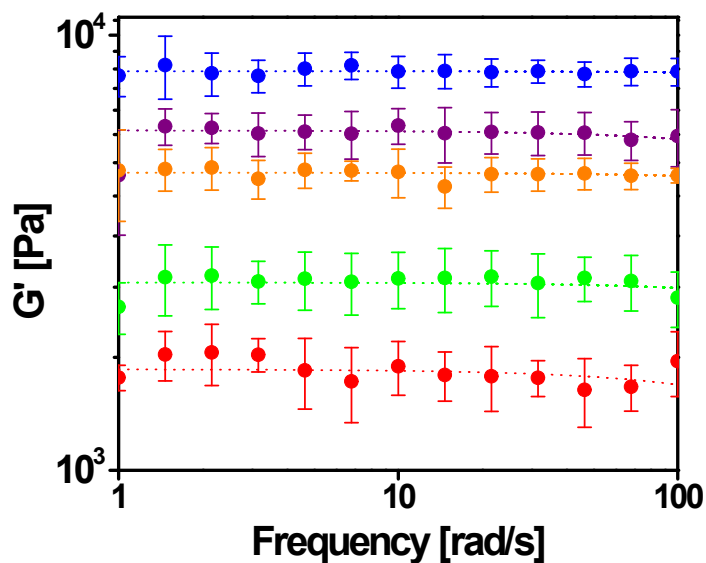
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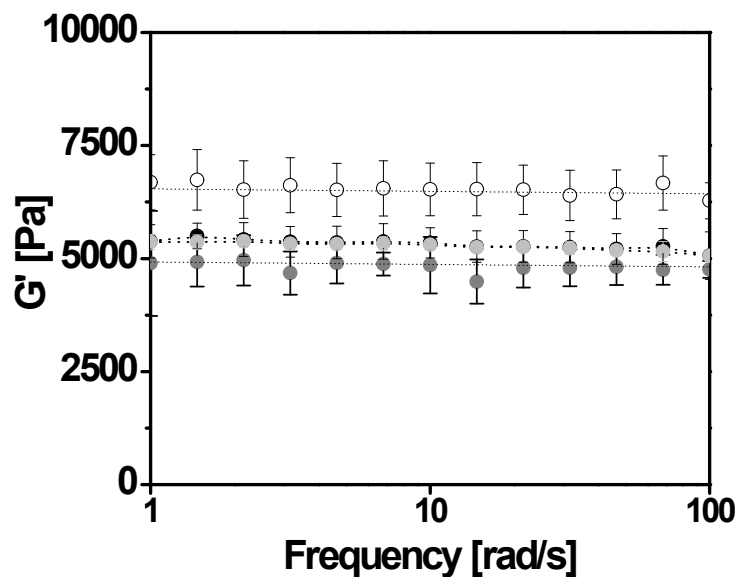
**Figure S1.** Correlation between the equilibrium mass-based swell ratio ( $Q_m$ ) and the weight fraction of high LCST precursor in the hydrogel. Swelling measured in 10mM PBS. Correlations: (○) 22°C  $Q_m = 6.62 \pm 0.54 + (0.081 \pm 0.011) \cdot x$ ;  $R^2 = 0.932$  and (●) 37°C  $Q_m = 3.68 \pm 0.43 + (0.090 \pm 0.007) \cdot x$ ;  $R^2 = 0.980$ .



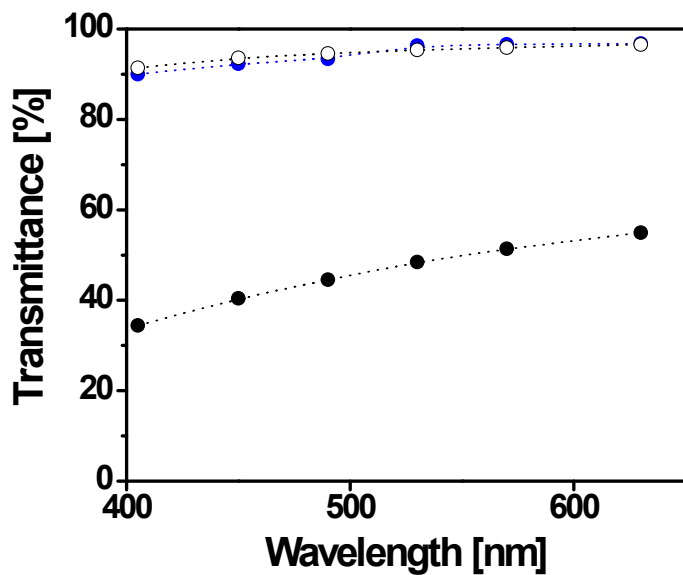
**Figure S2.** Swelling kinetics of PO(50/50) and PO(50/50) analogue hydrogels (with the same overall M(EO)<sub>2</sub>MA:OEGMA<sub>475</sub> ratio but a different distribution of the comonomers between the precursor polymer(s)) in 10 mM PBS: (●, orange) PO(50/50), (●, yellow) PO(L/H); (●, grey) PO(H/L) and (●, violet) PO<sub>55</sub>. Top figure: swelling kinetics measured at 22°C; bottom figure: swelling kinetics measured at 37°C.



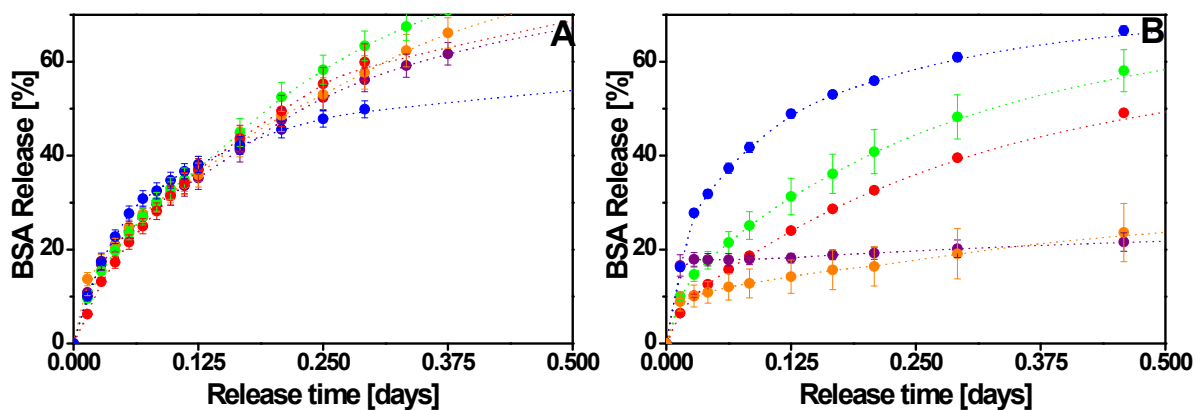
**Figure S3.** Mechanical properties of POEGMA hydrogels following swelling to equilibrium in 10 mM PBS at 22°C: (⊗, blue) PO(100/0); (⊗, purple) PO(75/25); (⊗, orange) PO(50/50); (⊗, green) PO(25/75) and (⊗, red) PO(0/100)



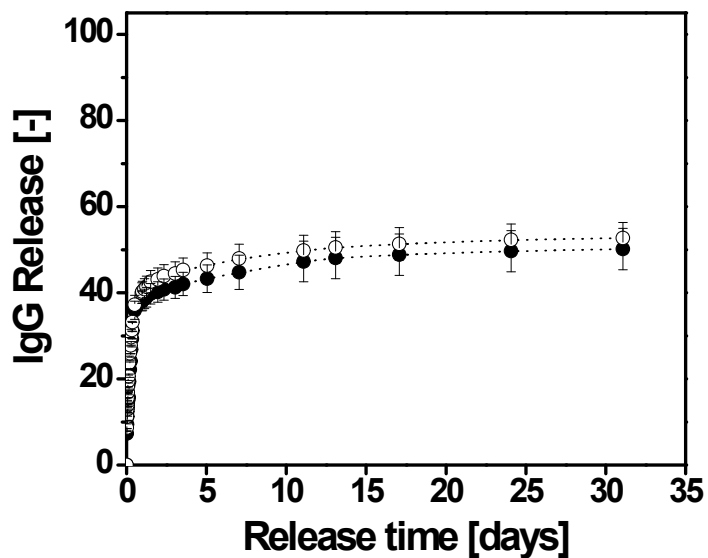
**Figure S4.**  $G'$  values for the PO(50/50) hydrogel as well as the PO(50/50) analogue hydrogels with the same overall  $M(\text{EO})_2\text{MA}:\text{OEGMA}_{475}$  ratio but a different distribution of comonomers between the precursor polymer(s). PO(50/50) (⊗, dark grey), PO55 (⊗, light grey), PO(L/H) (⊗, white) and PO(H/L) (⊗, black).



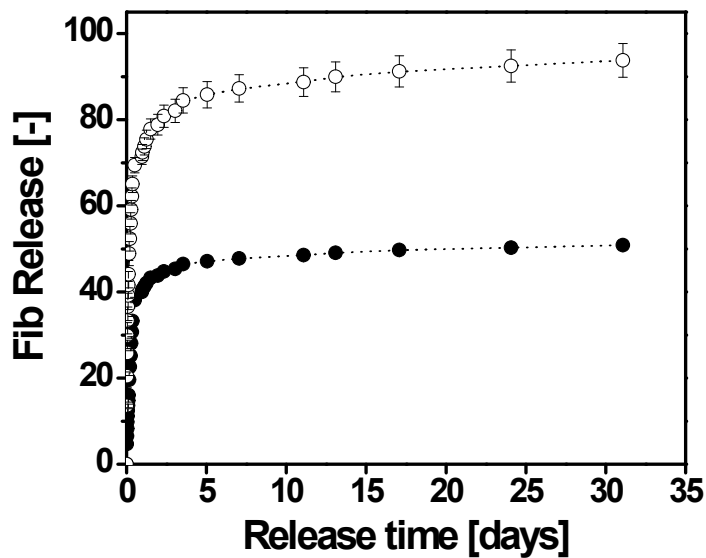
**Figure S5.** Optical transparency of PO(0/100) (●, black), PO(100/0) (○, open) and PO<sub>55</sub> (●, blue) as measured by UV-Vis at 37°C. Note that the lower transparency measured for PO(0/100) is due to the lower VPTT of this hydrogel and not because of phase separation between the precursors.



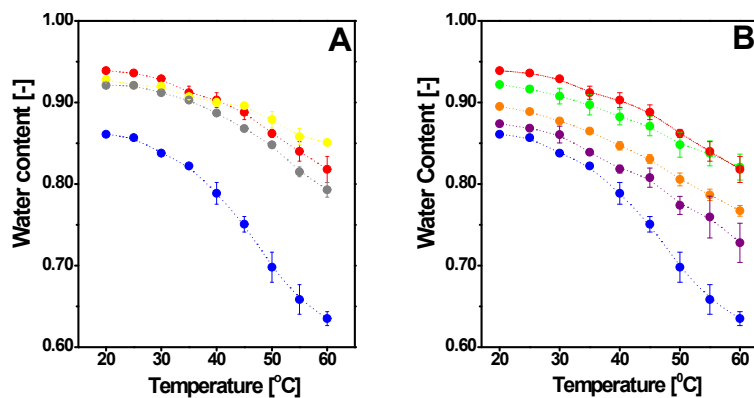
**Figure S6.** BSA release kinetics in 10mM PBS over the initial 12 hours of release at 37°C for the POEGMA hydrogels prepared at (A) 22°C and (B) 37°C: (●, blue) PO(100/0); (●, purple) PO(75/25); (●, orange) PO(50/50); (●, green) PO(25/75) and (●, red) PO(0/100).



**Figure S7.** Release of immunoglobulin G in 10mM PBS from PO(0/100) (●, black) and PO(100/0) (○, open) at 37°C.



**Figure S8.** Release of fibrinogen in 10mM PBS from PO(0/100) (●, black) and PO(100/0) (○, open) at 37°C.



**Figure S9.** Equilibrium water content of the mixed precursor POEGMA hydrogels as a function of the temperature in 10mM PBS: (A) single precursor and (B) mixed precursor hydrogels. PO<sub>0</sub> (●, black), PO<sub>10</sub> = PO(100/0) (●, blue), PO(75/25) (●, green), PO(50/50) (●, orange), PO(25/75) (●, purple), PO<sub>100</sub> (●, red), PO<sub>10</sub>H<sub>30</sub> + PO<sub>100</sub>A<sub>30</sub> (●, yellow) and PO<sub>100</sub>H<sub>30</sub> + PO<sub>10</sub>A<sub>30</sub> (●, grey).