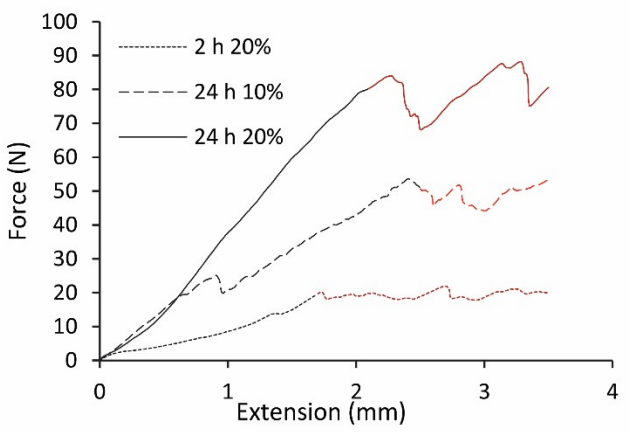
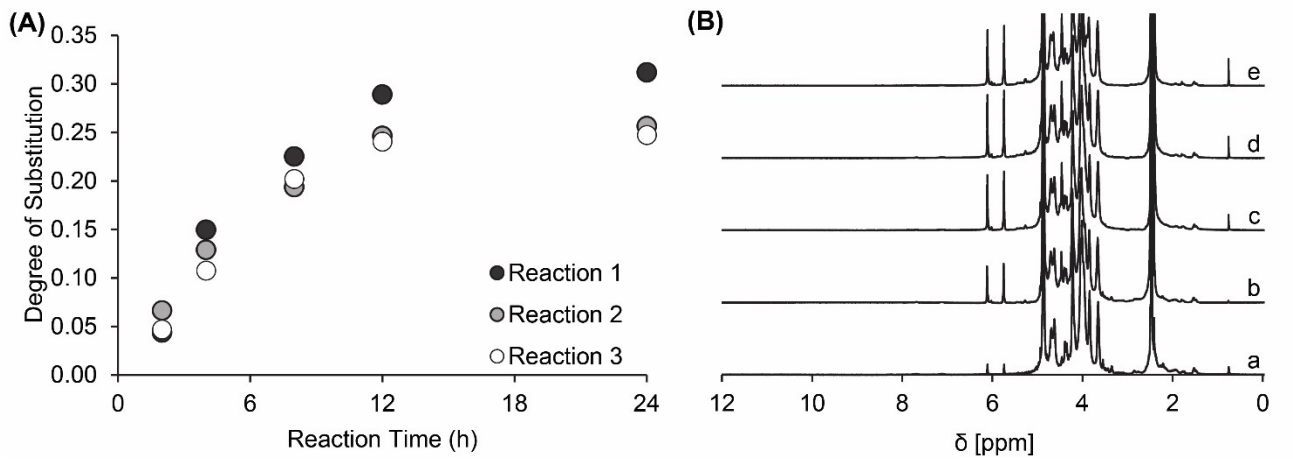
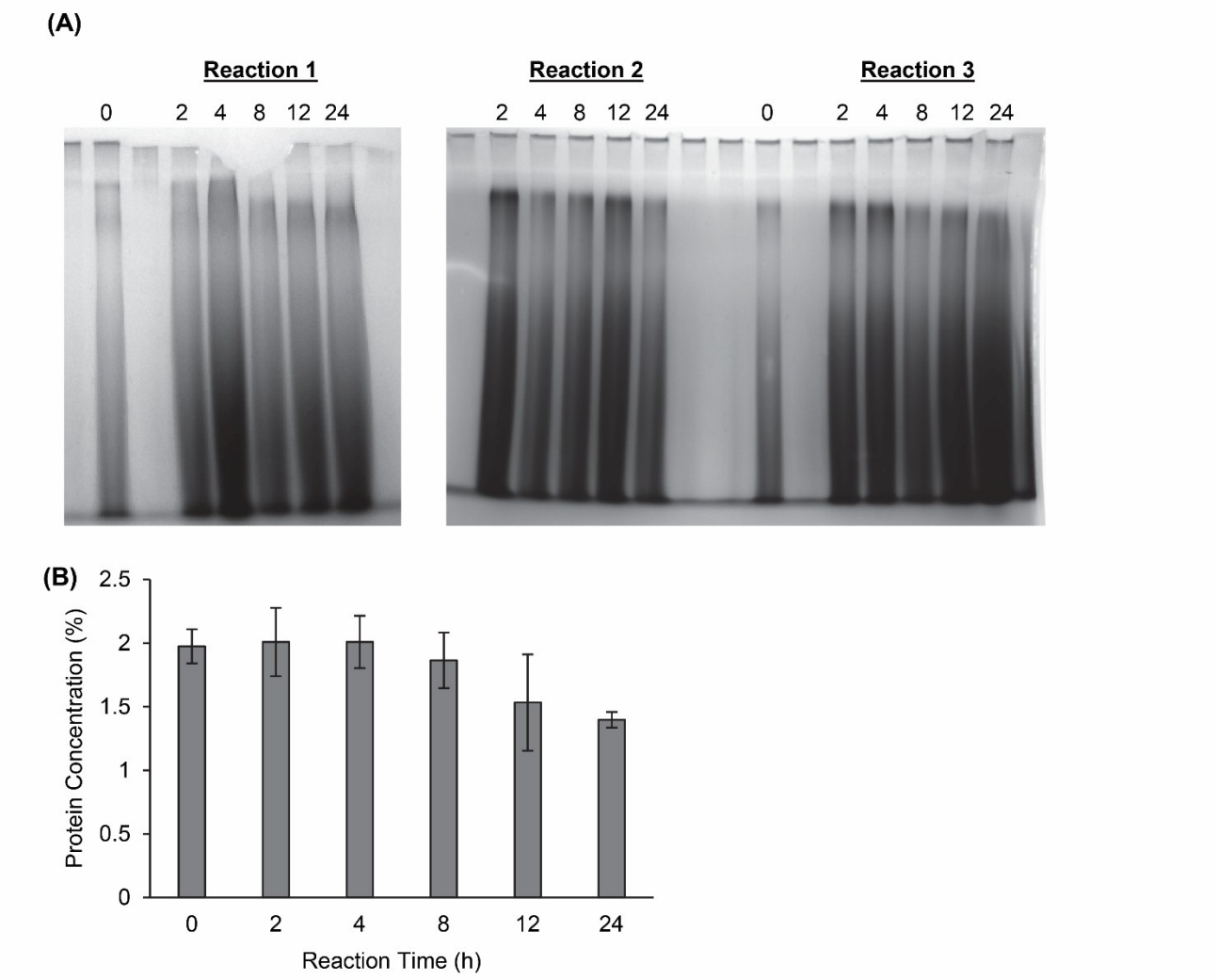
Electronic Supplementary Material (ESI) for Journal of Materials Chemistry B.

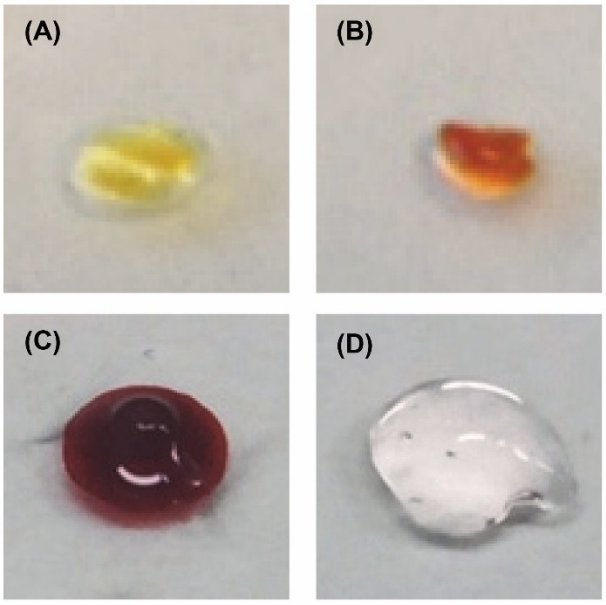

**Supplementary Figure 1. Injection force graphs of CSMA.** Representative injection force graphs of CSMA hydrogels. Hydrogels were injected for an extension distance of 3.5 mm at a rate of 4 mm/min. The plateau of the curve (shown in red) was used to calculate the average injection force.



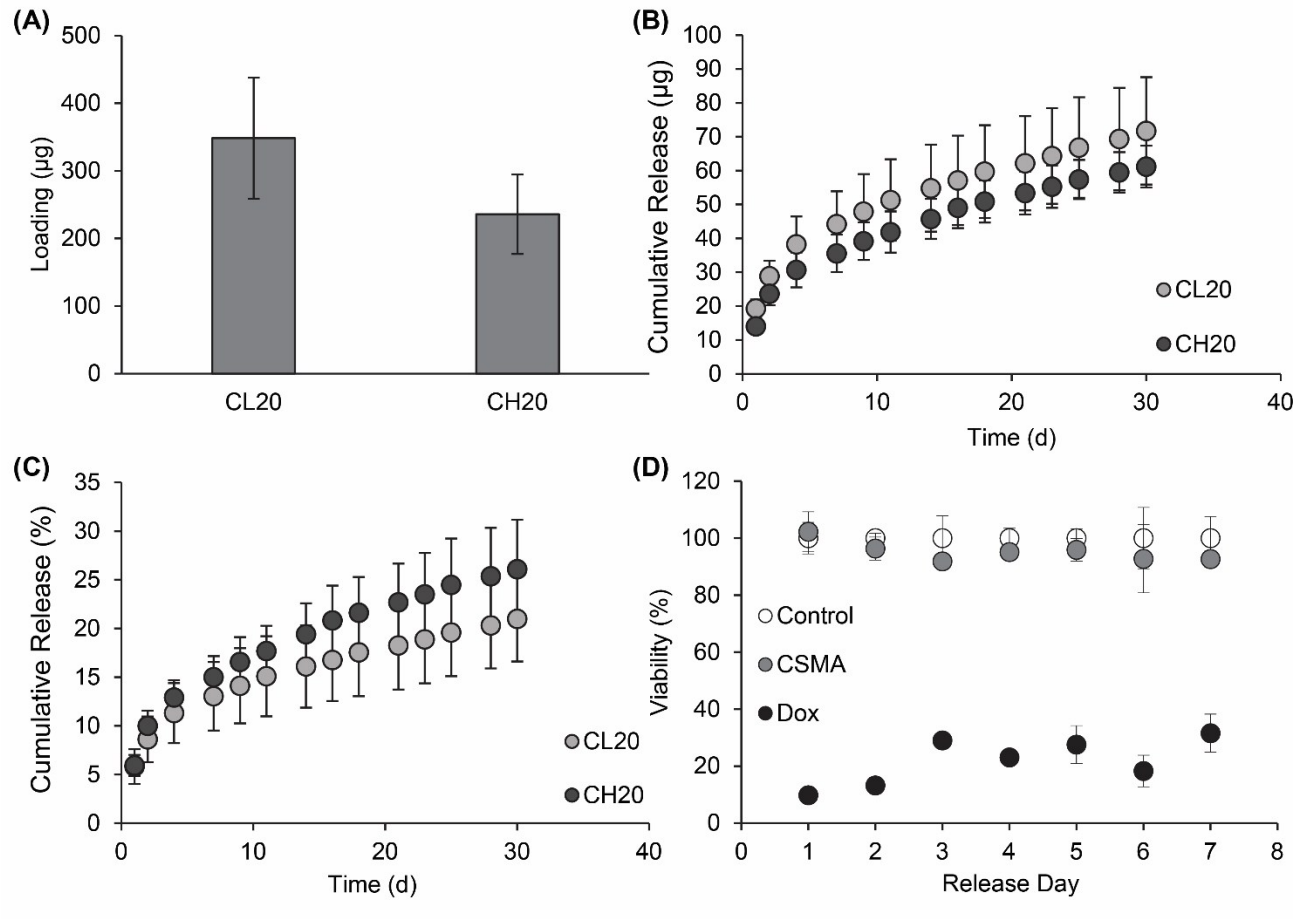

**Supplementary Figure 2.** H1 NMR analysis of CSMA. (A) Quantification of methacrylol peaks to determine the change in degree of substitution based on reaction time normalized to an alternate part of the CS backbone. (B) Full H1 NMR spectrum of CSMA reacted for (a) 2 h, (b) 4 h, (c) 8 h, (d) 12 h, and (e) 24 h.



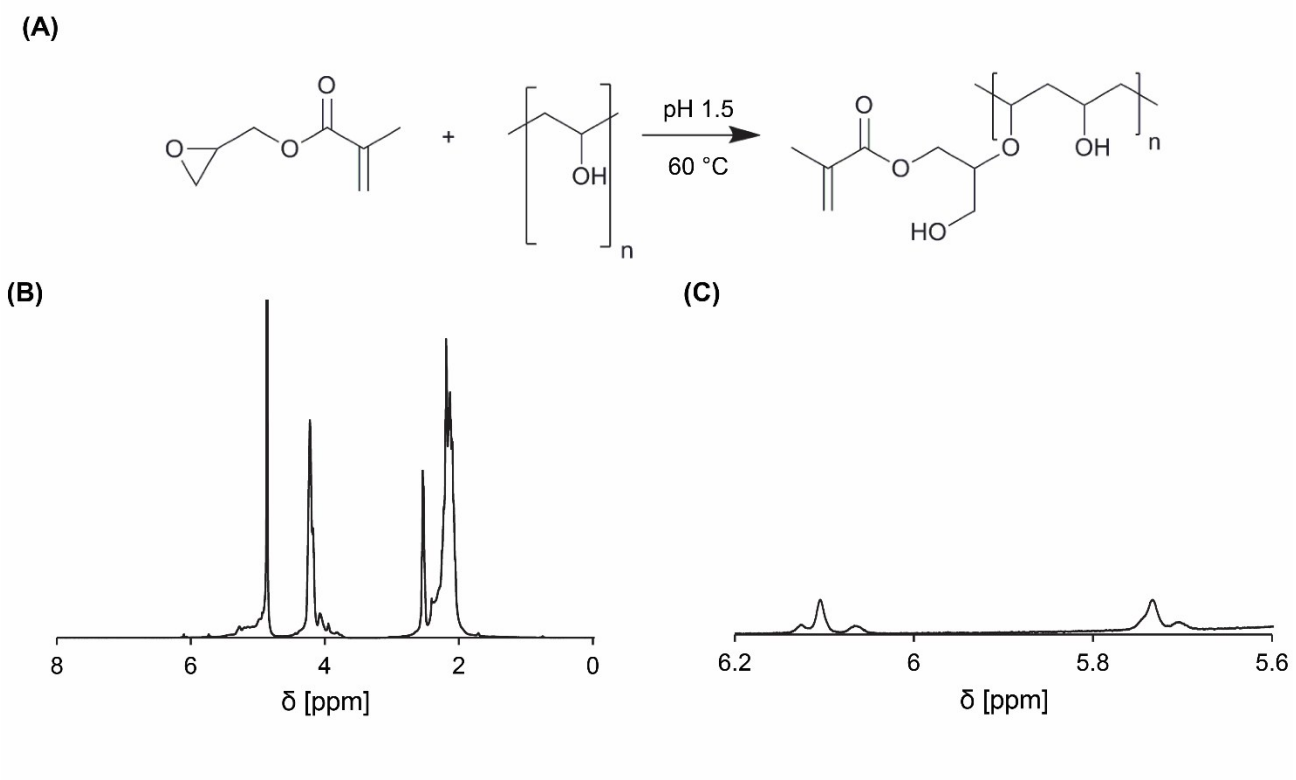

**Supplementary Figure 3.** Protein and molecular weight characterization of CSMA. (A) CSMA resolved on 4-15% Polyacrylamide gels and stained with 0.5% alcian blue. (B) Protein concentration (%) after methacrylation of CS at each time point (h).



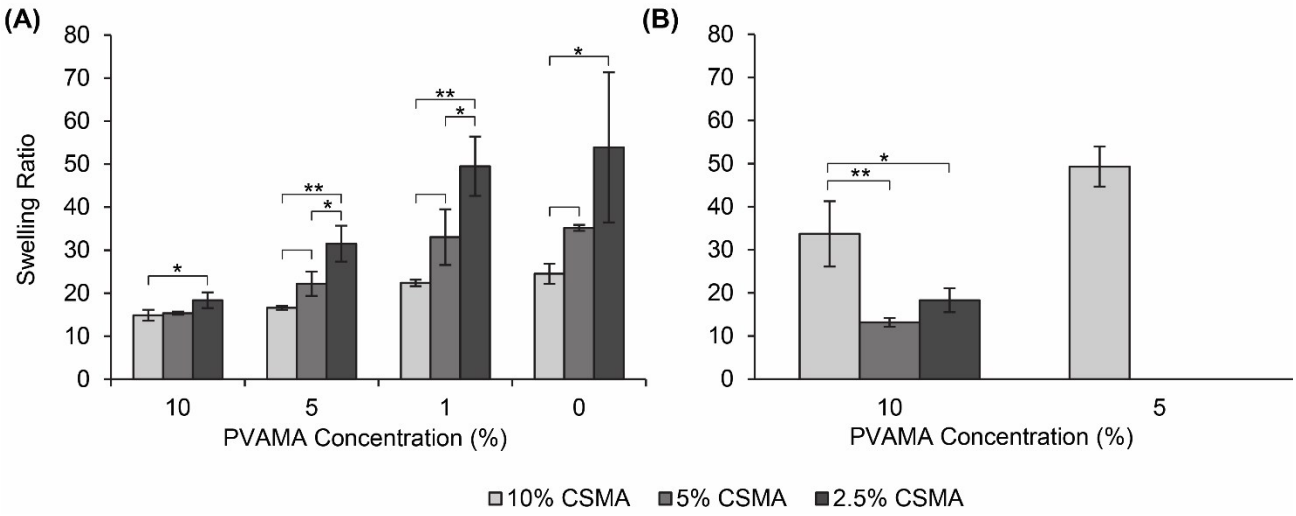

**Supplementary Figure 4.** Images of hydrogels post-drug release. (A) SUN-loaded CL20 hydrogel after 21 days of release. (B) SUN-loaded CH20 hydrogel after 21 days of release. The difference in color intensity from light yellow/orange to deep orange pictorially shows the difference in remaining drug from low to high, respectively. (C) DOX loaded CH20 hydrogel after 7 days of release. (D) DOX loaded 20% (w/v) PVAMA hydrogel after 7 days of release. The lack of red color shows no DOX remaining, any initial DOX in the PVAMA hydrogels is a function of solution equilibration inside and outside of the hydrogel – not drug binding.



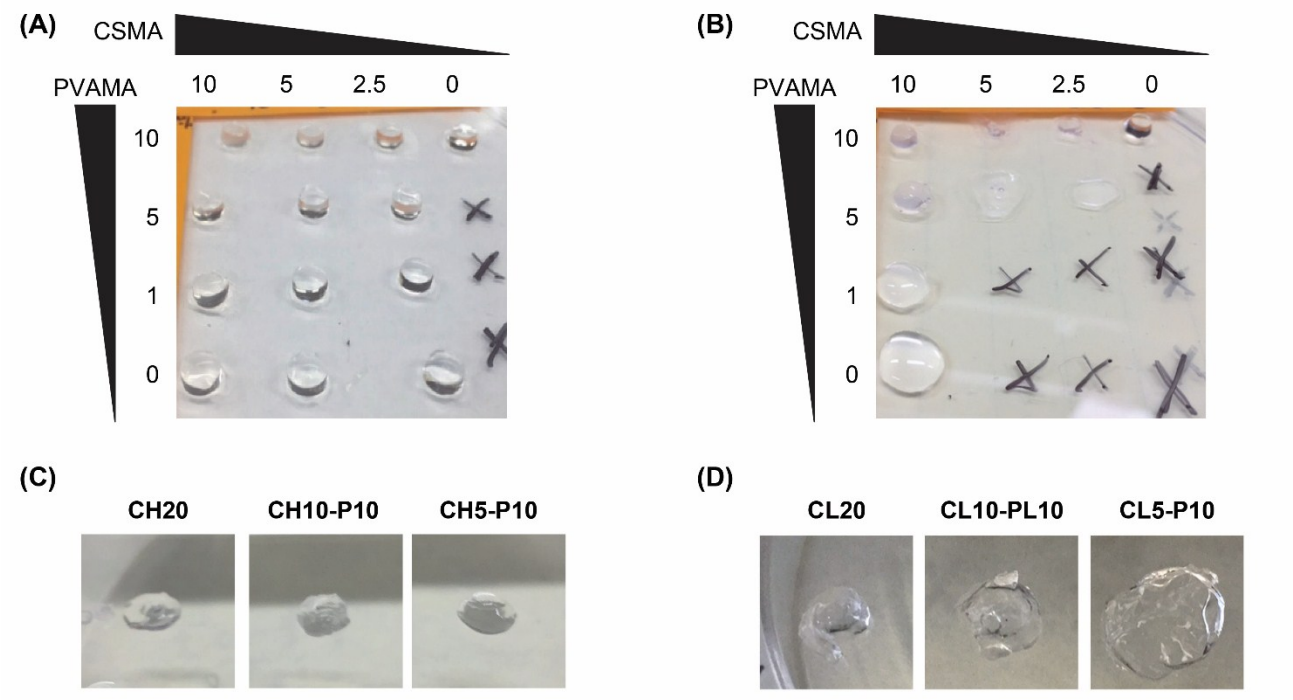

**Supplementary Figure 5.** Impact of DS on DOX loading and release and evaluation of release cytotoxicity *in vitro*. 20% (w/v) CL20 and CH20 hydrogels were evaluated for (A) Mass loading of DOX. (B) Cumulative mass release of DOX. (C) Cumulative percent release of DOX. (D) Direct cytotoxicity of SUN loaded hydrogels on KELLY neuroblastoma cells. (A-C) Data are presented as mean ± standard deviation of three independent experiments. (D) Data are presented as mean ± standard deviation of three independent samples.




**Supplementary Figure 6. Synthesis of PVAMA.** (A) Glycidyl methacrylate was reacted with poly(vinyl alcohol) for 24 h to synthesis PVAMA. (B) $H^1$ NMR spectrum of PVAMA. (C) $H^1$ NMR spectrum in region of vinyl protons on methacrylate at δ 6.11 and δ 5.74 ppm.



**Supplementary Figure 7. Swelling ratio of CSMA/PVAMA composite hydrogels**. (A) Swelling ratio of CSMA/PVAMA composite hydrogels fabrication with CSMA reacted for 24 h (high DS, 0.28). (B) Swelling ratio of CSMA/PVAMA composite hydrogels fabrication with CSMA reacted for 2 h (low DS, 0.05). Data are presented as mean ± standard deviation of three independent experiments. Asterisks denotes statistical differences between indicated groups ($^{*}p < 0.05$, $^{**}p < 0.01$, $^{***}p < 0.001$).




**Supplementary Figure 8.** Images of composite CSMA/PVAMA hydrogel. Images of pre-swelling hydrogels fabricated from CSMA/PVAMA composites using (A) high DS CSMA and (B) low DS CSMA fabricated in PBS. Images of post-swelling hydrogels fabricated from CSMA/PVAMA composites using (C) high DS CSMA and (D) low DS CSMA fabricated in water. Hydrogel formulations in (C) and (D) were used for the SUN release studies.