Electronic Supplementary Information for

Self-healing Hydrogels Containing Reversible Oxime Crosslinks

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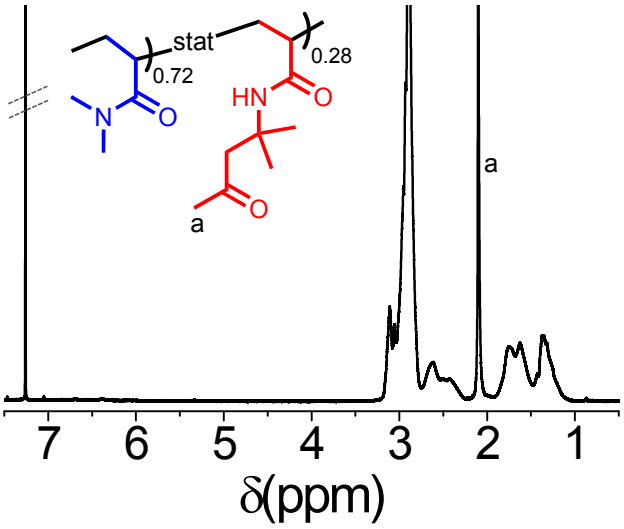

Figure S1. ^{1}H NMR spectrum of poly(*N,N*–dimethylacrylamide-*stat*-diacetone acrylamide) (P(DMA$_{0.72}$-*stat*-DAA$_{0.28}$)).

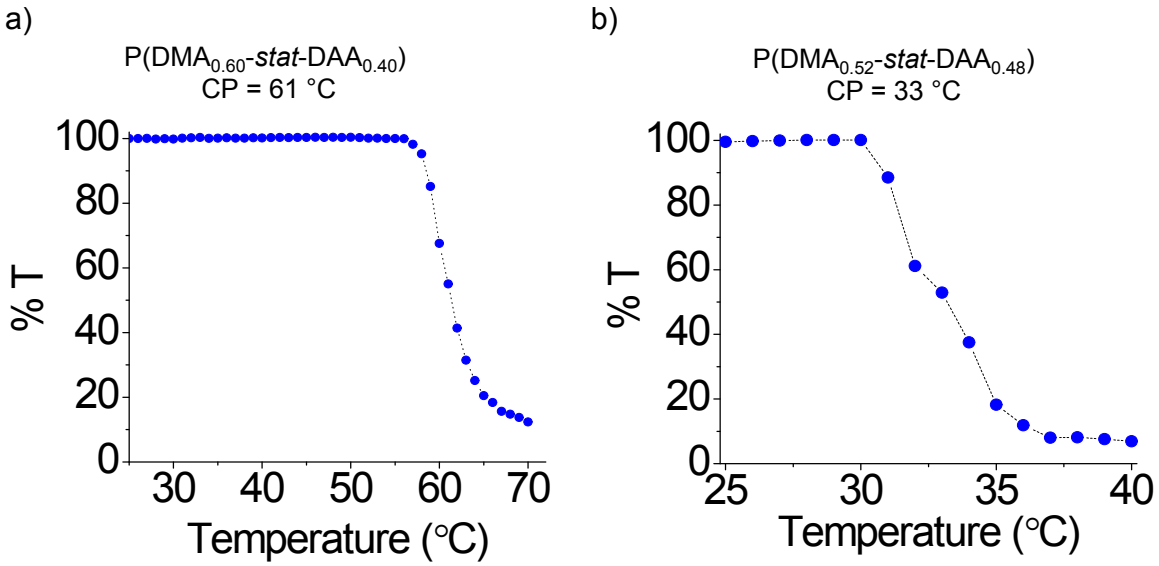

Figure S2. Determination of cloud point (CP) *via* turbidity by monitoring the change in transmission (%T) with increasing temperature of (a) P(DMA$_{0.6}$-*stat*-DAA$_{0.4}$) (b) P(DMA$_{0.52}$-*stat*-DAA$_{0.48}$) (0.5 mg/mL in deionized water).

Figure S3. Reversible sol-to-gel transition of P($DMA_{0.60}$–*stat*–$DAA_{0.40}$) (a) by vial inversion and (b) by temperature step rheology experiments over 3 cycles at 10 rad/sec and 1% strain.

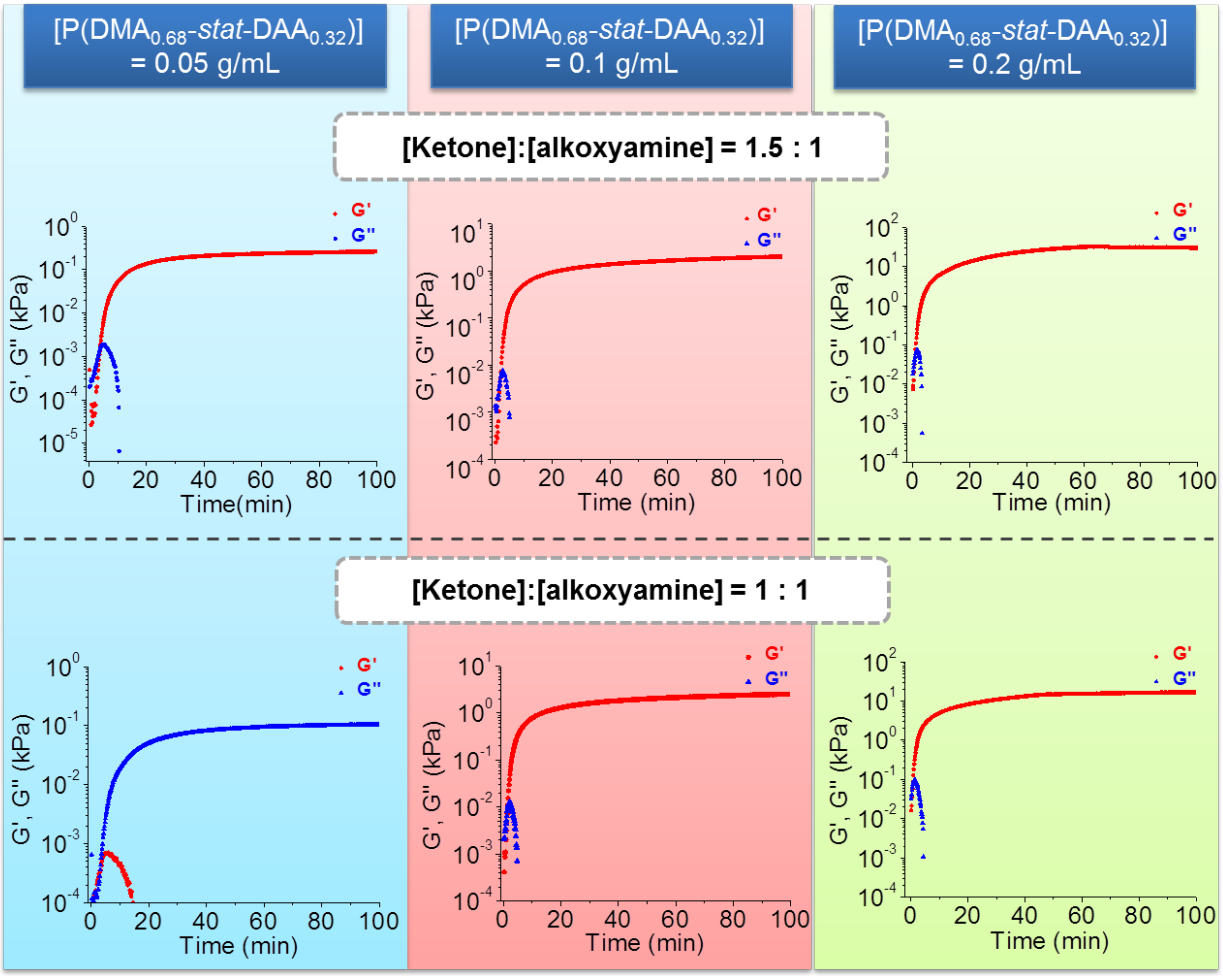

Figure S4. Dynamic time sweep experiments for hydrogels formed from P($DMA_{0.68}$–*stat*–$DAA_{0.32}$) at different concentrations (*i.e.,* 0.05, 0.1, and 0.2 g/mL) and different [ketone]:[alkoxyamine] ratios (*e.g.,*1.5:1 and 1:1).

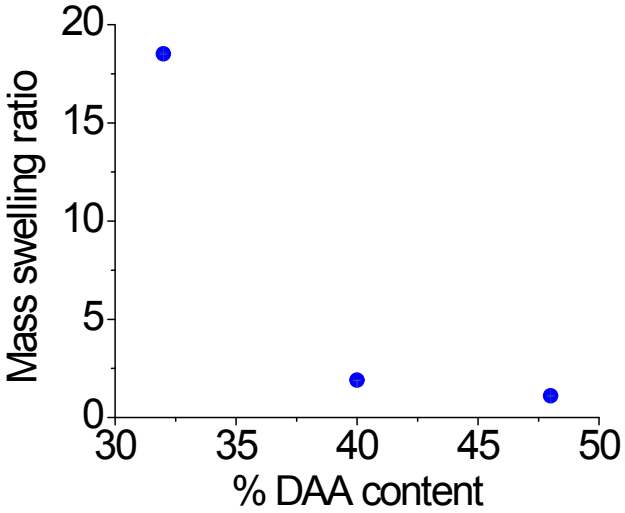

Figure S5. Mass swelling ratio of hydrogels with different DAA content of P(DMA_m–*stat*–DAA_n). Mass swelling ratio = (Mass of swollen gel - Mass of dry gel)/Mass of dry gel.

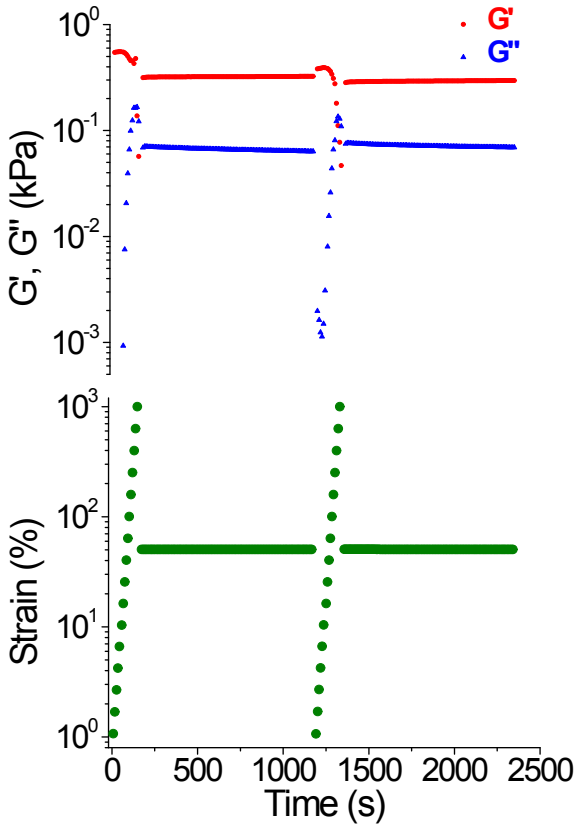

Fig. 6 Self-healing of hydrogels after fracture. The top plot shows the change in modulus (red and blue) during the strain ramp (green) described by the bottom plot. The hydrogel fractured during the strain ramp up to 630% and rapidly healed after the strain was reduced. The strain ramp was conducted at constant angular frequency of 10 rad/s and a constant temperature 25 °C. [P($DMA_{0.68}$-*stat*-$DAA_{0.32}$)] = 0.05 g/mL, and [ketone]:[alkoxyamine] = 1.5:1 using a 50 mm CP geometry.

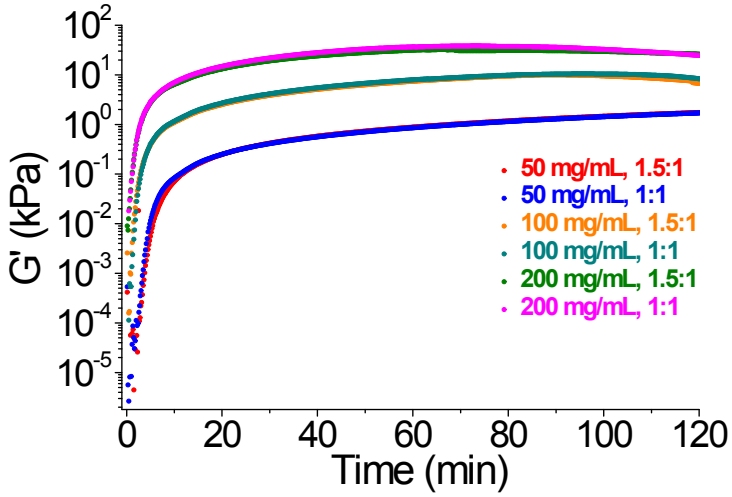

Figure S7. Evolution of elastic modus (G′) with time during time sweep experiments of hydrogels prepared with different polymer concentrations (*i.e.,* [P($DMA_{0.60}$–*stat*–$DAA_{0.40}$)] =0.05, 0.1, 0.2 g/mL) and stoichiometries (*i.e.*, [ketone]:[alkoxyamine]=1.5:1 and 1:1).

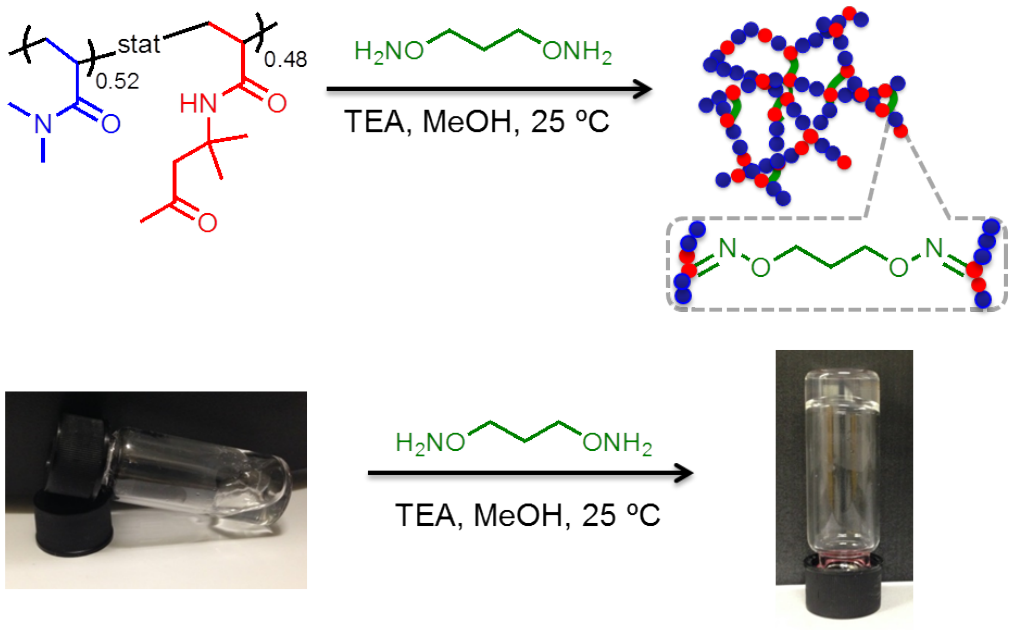

Figure S8. Preparation of organogel at [P($DMA_{0.52}$-*stat*-$DAA_{0.48}$)] = 200 mg/mL, [ketone]:[alkoxyamine] = 1.5:1 in methanol.

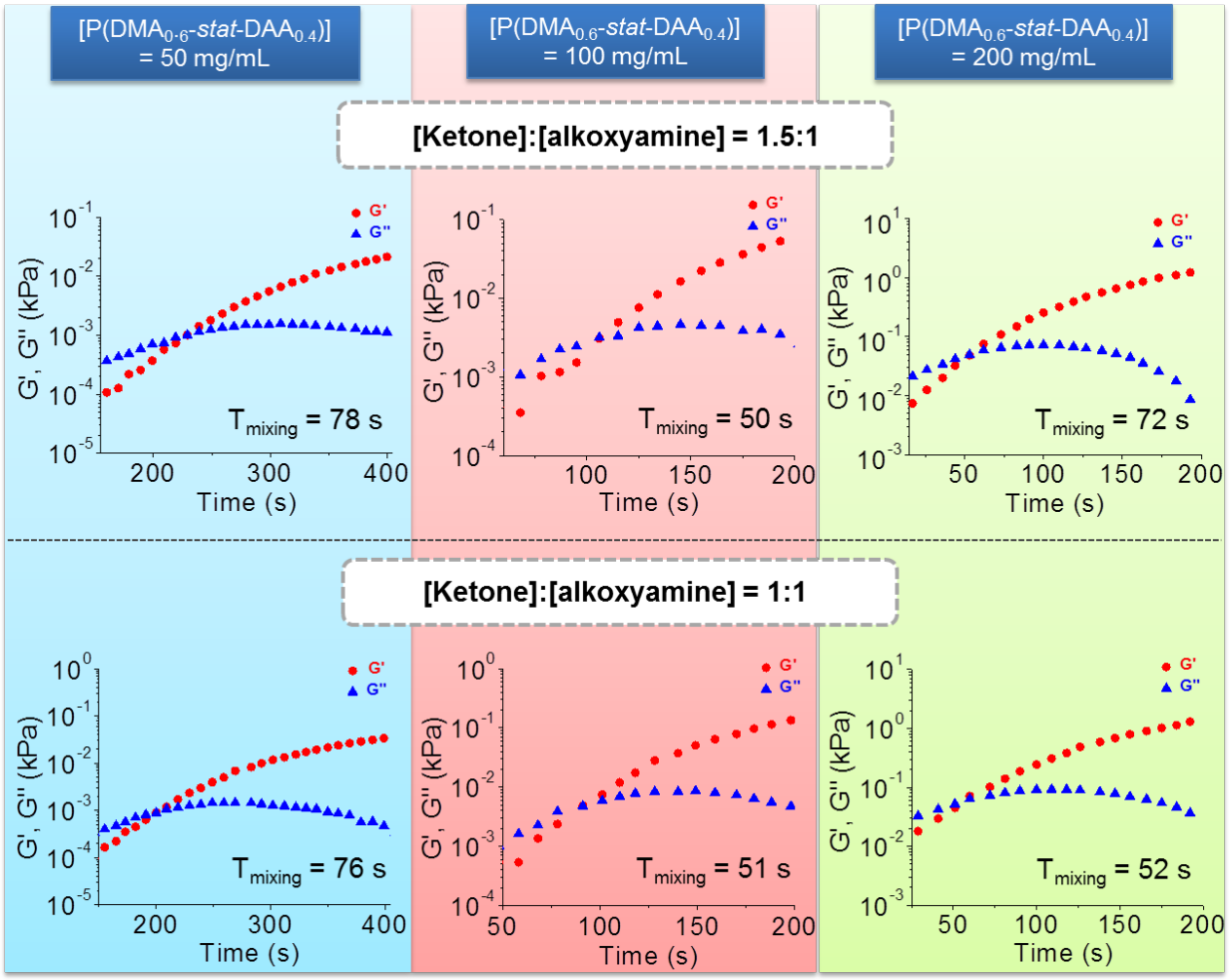

Figure S9. Dynamic time sweep experiments for hydrogels formed from P($DMA_{0.6}$–*stat*–$DAA_{0.4}$) at different concentrations (*i.e.,* 0.05, 0.1, and 0.2 g/mL) and different [ketone]:[alkoxyamine] ratios (*e.g.,*1.5:1 and 1:1).