Supporting Information

Adhesion between Hydrophobic Elastomer and Hydrogel through Hydrophilic Modification and Interfacial Segregation

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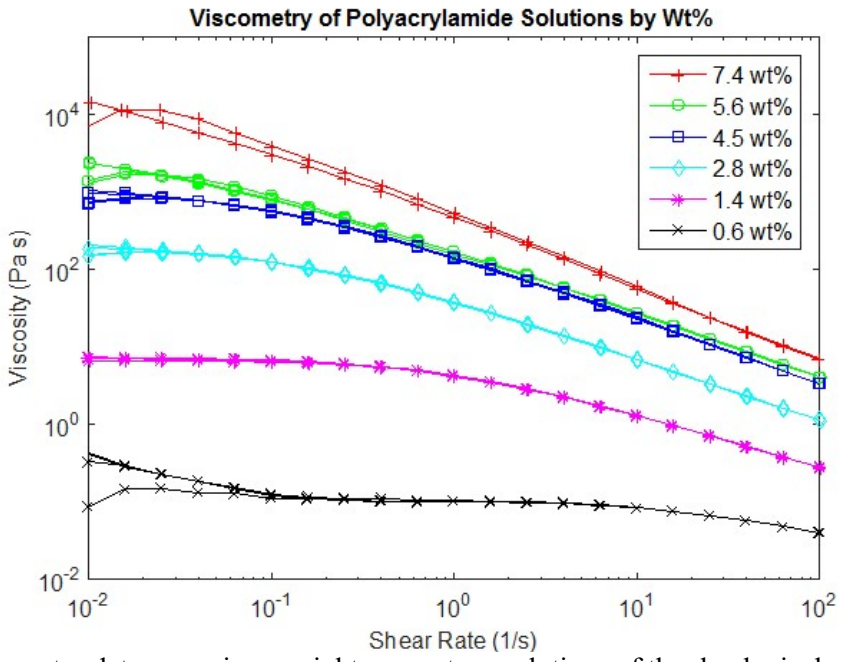

Figure S1. Viscometry data on various weight-percentage solutions of the rheological modifier polyacrylamide. A previous study utilized a 7.4wt% PAAm solution, whereas this study used an equivalent 5.6wt% PAAm solution. Our internal tests suggest that extrusion printing with solutions as low as 2.8 wt% are possible.

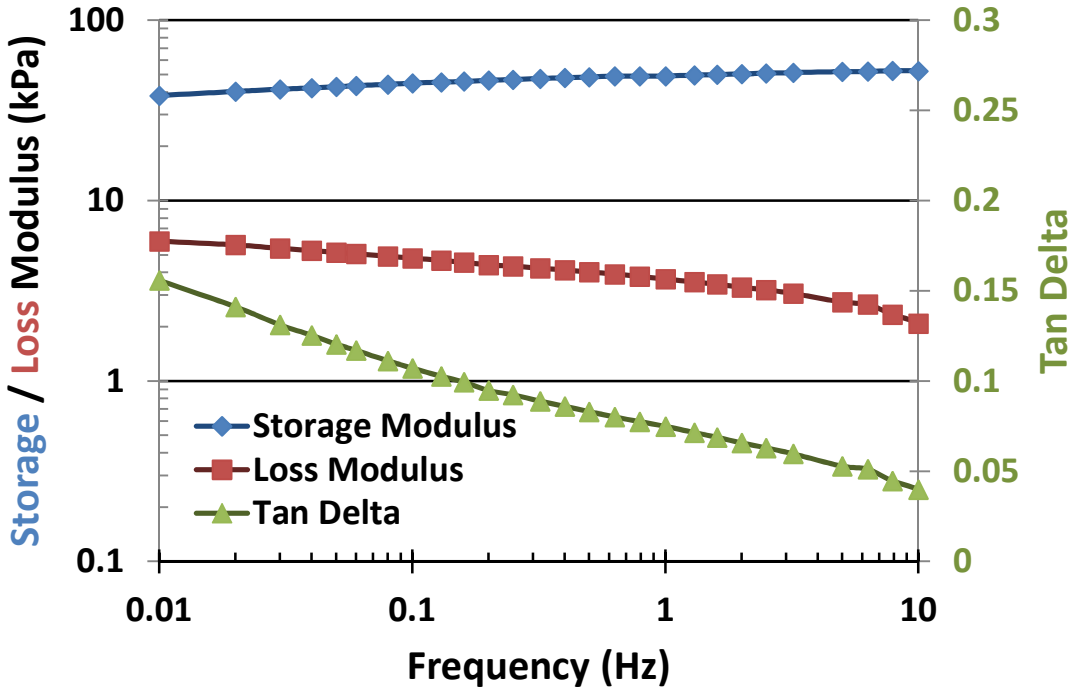

Figure S2. Dynamic Mechanical Analysis (DMA) data on bulk hydrogel containing rheological modifier using a DMA Q800 (TA Instruments). The test was performed under compression at maximum oscillatory strain of 3% on a cylindrical sample at 25°C and an initial contact force of 0.01N.

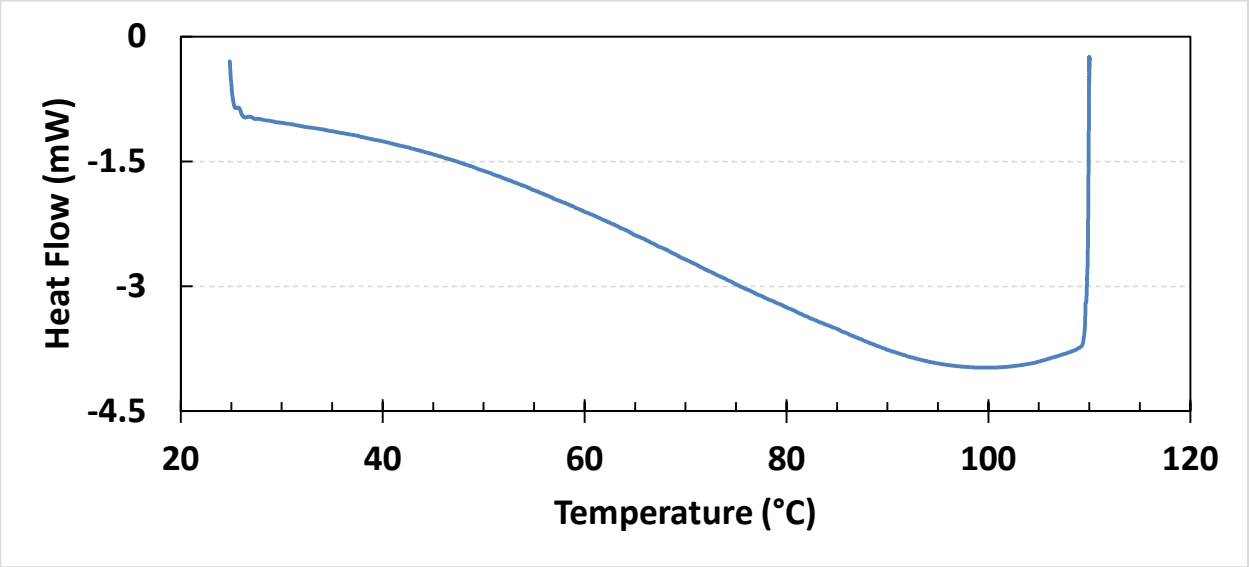

Figure S3. Differential Scanning Calorimetry (DSC) measurement of a bulk hydrogel containing rheological modifier, with endothermic being negative on this scale. Measurements made on a DSC Q200 (TA Instruments). A heating sweep from 25°C to 110°C is made and we notice only a broad peak centered around 100°C. Samples were contained in a Tzero Low Mass Aluminum pan. The DSC was operated under a 40 µL min−1 nitrogen flow rate, 5 C°/min temperature ramp and data captured at a rate of 1 Hz.

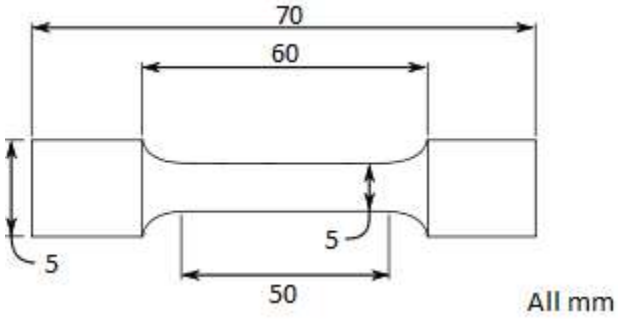

Figure S4: Dogbone geometry used for stress relaxation hydrogels tests. All units in mm.

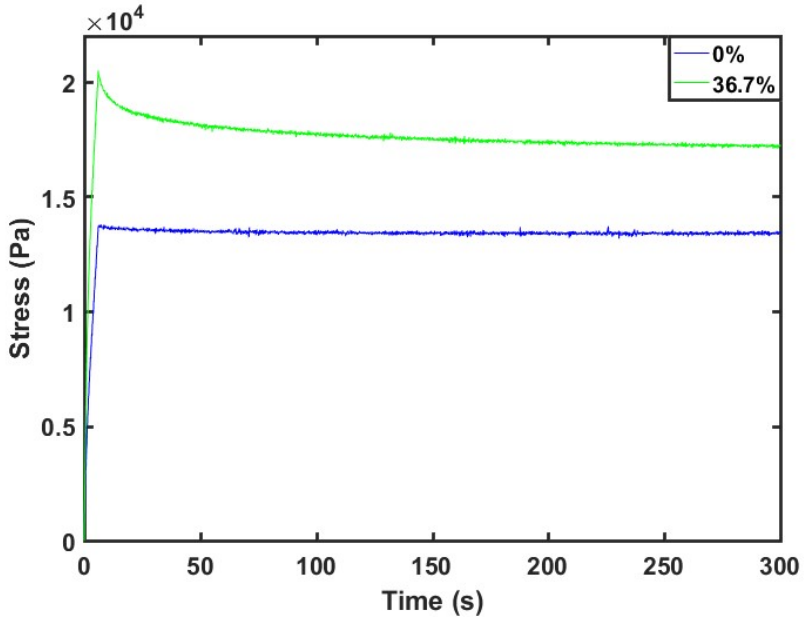

Figure S5. Stress-relaxation tensile testing of bulk hydrogel samples with varying ratios of uncrosslinked to crosslinked PAAm within the network. This illustrates the small effect of uncrosslinked PAAm on the bulk hydrogel mechanical properties.

SI Video 1: A bilayer shear test sample of hydrogel containing rheological modifier and plasma-treated PDMS aged for 4 days.

SI Video 2: A bilayer sample with hydrogel without rheological modifier and plasma-treated PDMS aged for 14 days.

SI Video 3: A bilayer sample with hydrogel containing rheological modifier and plasma-treated PDMS aged for 17 days.