Supporting Information

Patterned, Morphing Composites via Maskless Photo-click Lithography

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FTIR-ATR spectra were collected with repeated scan times of 64 and a resolution of 4 cm⁻¹. The appearance of peak at 944 cm⁻¹ in the chitosan-MA curve results from the out of plane deformation of –C=CH₂ in the methacrylamide structure. After functionalization, the increased absorbance of the peak at 1548 cm⁻¹ and range of 560-650 cm⁻¹ come from the amide II band (N-H bending and C-N stretching vibrations) and the bending motion of O=C–N, respectively.

Figure S1: FTIR spectra for chitosan and chitosan-MA.

¹H NMR spectroscopy was conducted on a Bruker 500 MHz at 25 °C. Chitosan and chitosan-MA (0.5% (w/v)) were dissolved in 0.5% deuterium chloride (DCl) in deuterium oxide (D₂O) at room temperature. The appearance of the proton peaks at 5.28, 5.6 ppm and 1.7-1.9 ppm results from
the –C=CH₂ and –C–CH₃ groups on the methylene binding of the chitosan-MA. The methacrylation degree is determined by the integrated area ratio between the –C=CH₂ of the methylene group at 5.28-5.6 ppm and the H3-H6 peaks of chitosan at 3.3-4.0 ppm².³

Figure S2: ¹H NMR spectra for chitosan and chitosan-MA
Figure S3: Raman Spectroscopy of GO and rGO

Figure S4: $^1$H NMR spectrum for the photoinitiator, LAP
Figure S5: UV-visible spectrum for the photoinitiator, LAP
Figure S6: (a) Water-in-air (w/a) contact angle of PDMS surface before plasma. (b-d) w/a contact angle of PDMS surface after plasma treatment in 0 min, 20 min and 120 min, respectively. (e) XPS spectrum of thiolated PDMS (Inset: Deconvoluted XPS of S 2s for thiolated PDMS), the percentage of S 2s is 1.57 atom% on the thiolated PDMS surface. (Due to the overlap issue between primary S 2p and Si 2s in the spectrum, Sulphur content was characterized by S 2s in this study.)
Figure S7: Demonstration of the elongated behaviours during the tensile test for covalent bonded bilayered composite and non-covalent bonded bilayered composite. (a) Stretching process for covalent bonded bilayered composite. Only one major crack happened on the rGO-chitosan-MA layer before the failure of whole composite. (b) The failure point of the covalent bonded bilayered composite. Composite crack went through the rGO-chitosan-MA layer crack. (c) Stretching process for non-covalent bonded bilayered composite. The rGO-chitosan-MA layer split into few parts before PDMS layer cracked. (d) The point before failure of the non-covalent bonded bilayered composite. (Inset: the failure point of the non-covalent bonded bilayered composite. The sample curled back and caused observation difficulty, so the point before failure was used for demonstration.)
Figure S8: Relationship between the thickness of rGO-chitosan-MA layer and bending angle when PDMS thickness and temperature change remain the same on the basis of thermomechanics study of bimetallic thermostats in Equation (1).

REFERENCES

