Electronic Supplementary Material (ESI) for Soft Matter.

Supporting information for *Direct ink writing of tough, stretchable silicone composites*

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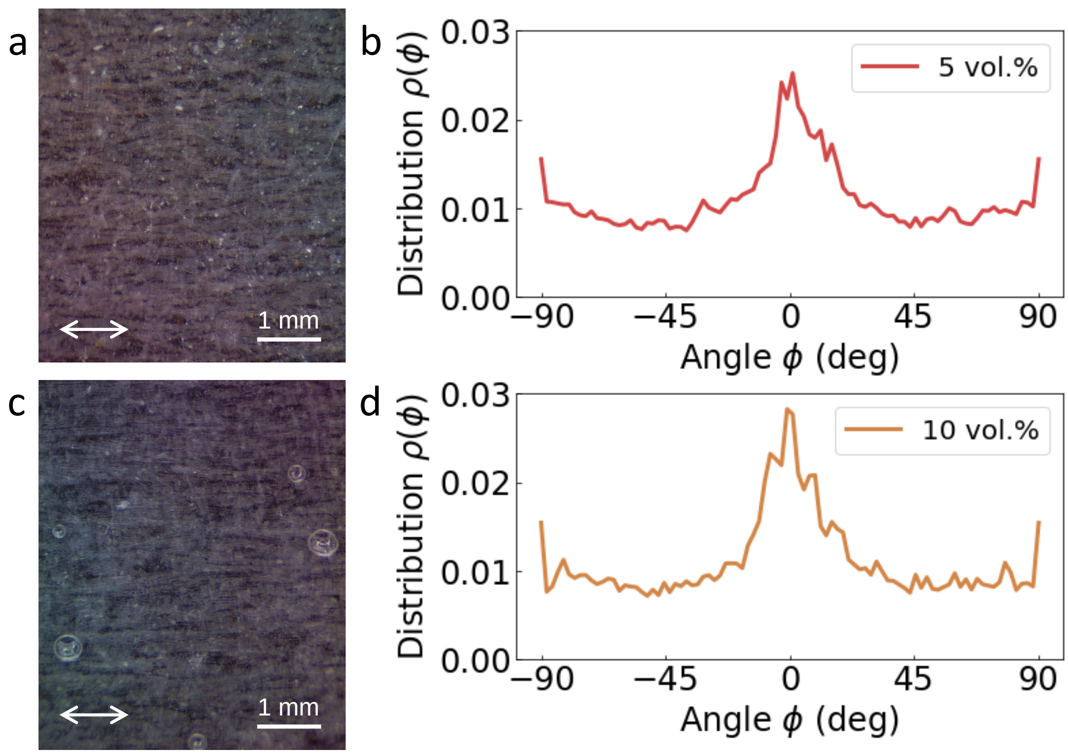

Figure S1: (a) Optical microscope image of surface of 5 vol.% PDUO-GF composites. (b) Distribution of fiber orientations for 5 vol.% composites. (c) Optical microscope image of surface of 10 vol.% PDUO-GF composites. (d) Distribution of fiber orientations for 10 vol.% composites. The fibers in the images are seen to be aligned in the direction of printing, as indicated by the white arrows.

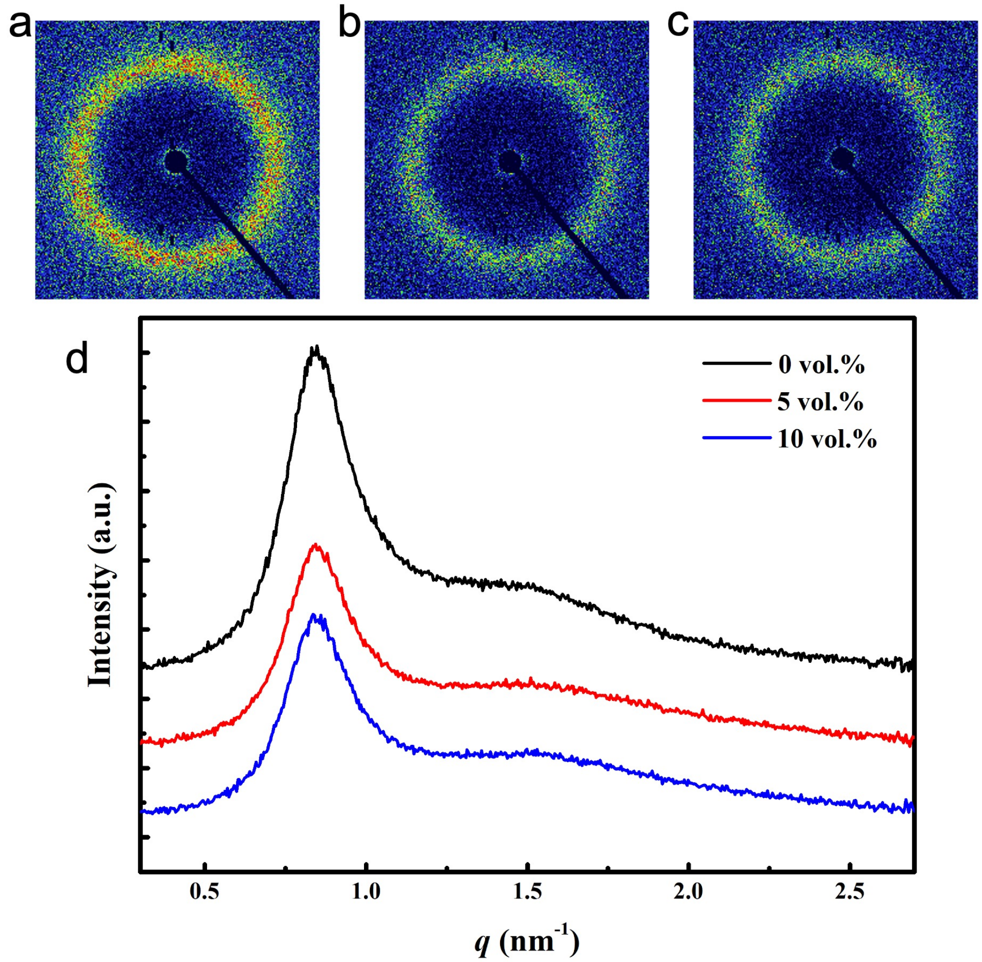

Figure S2: Small angle X-ray scattering (SAXS) patterns for 3D-printed PDUO with (a) 0 vol.%, (b) 5 vol.% , and (c) 10 vol.% glass fibers. (d) SAXS spectrum for 3D-printed PDUO. The SAXS results indicate the existence of hydrogen bonding interdomain with a distance of 7.4 nm. The presence of GFs does not significantly alter the spectrum. SAXS was performed using a Xeuss 2.0 Dual Source Environmental X-ray Scattering System with Cu source, 0.154 nm wavelength, at 50 kV and 0.6 mA current. The sample was in a vacuum chamber at room temperature. The sample-detector distance was set to 177 mm, giving a measurement 2θ value range of 0.92° to 37°. The X-ray exposure time was 600 s.

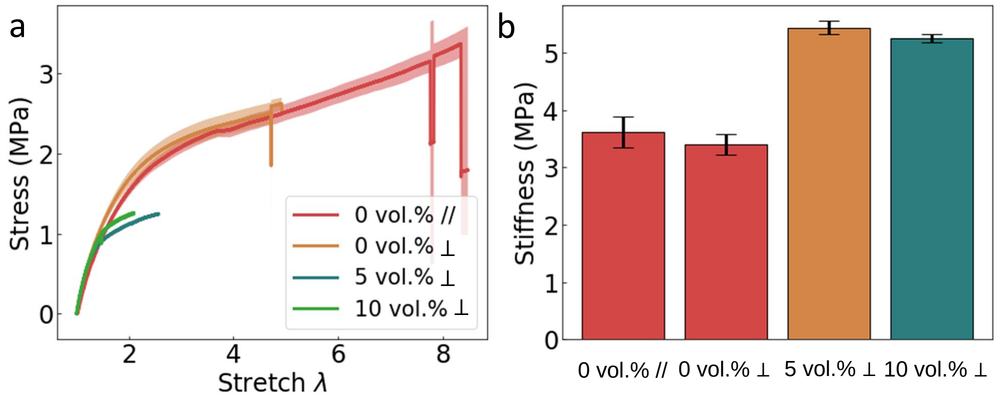

Figure S3: (a) Stress-stretch response of PDUO (with no fibers) when loaded parallel with or perpendicular to the print direction, and response of PDUO-GF composites when loaded perpendicular to the fiber (and print) direction. (b) Stiffness of 3D-printed PDUO and PDUO-GF composites when loaded perpendicular to the print path.

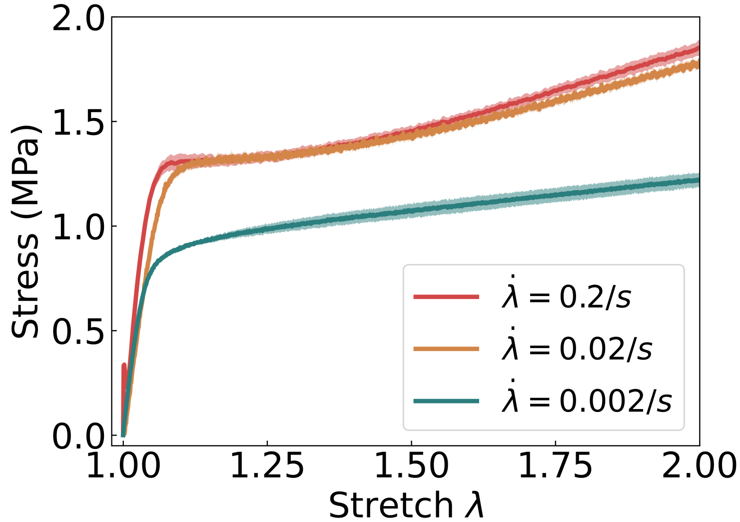

Figure S4: Stress-stretch responses of PDUO-GF composites at 5 vol.% when loaded at different loading rates. When the composite is loaded at a much slower speed, the overall stress is much lower at stretches beyond initial yielding.

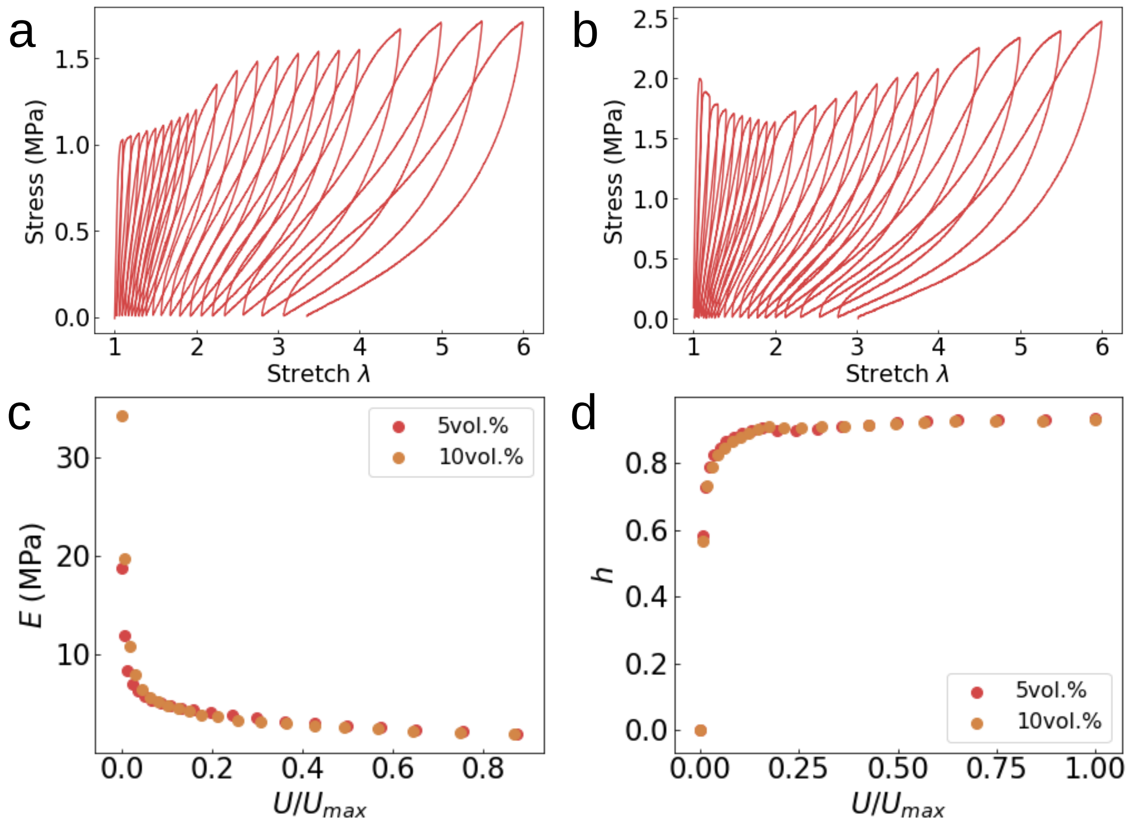

Figure S5: Cyclic loading of PDUO-GF composites: (a) and (b) Stress-stretch response of 5 vol.% and 10 vol.% composites, respectively, under cyclic loading of increasing stretch amplitude. (c) Initial stiffness during each reloading step as a function of strain energy density, normalized by the maximum strain energy density at fracture. (d) Hysteresis ratio as a function of normalized strain energy density, representing the degree of damage and fraction of energy dissipated during the loading and unloading process. The hysteresis ratio h indicates the fraction of strain energy that is dissipated during loading and unloading. It is defined as $h = W_D/W$, where W_D is the work dissipated during the loading-unloading cycle and W is the strain energy density of the pristine material at a given stretch λ.

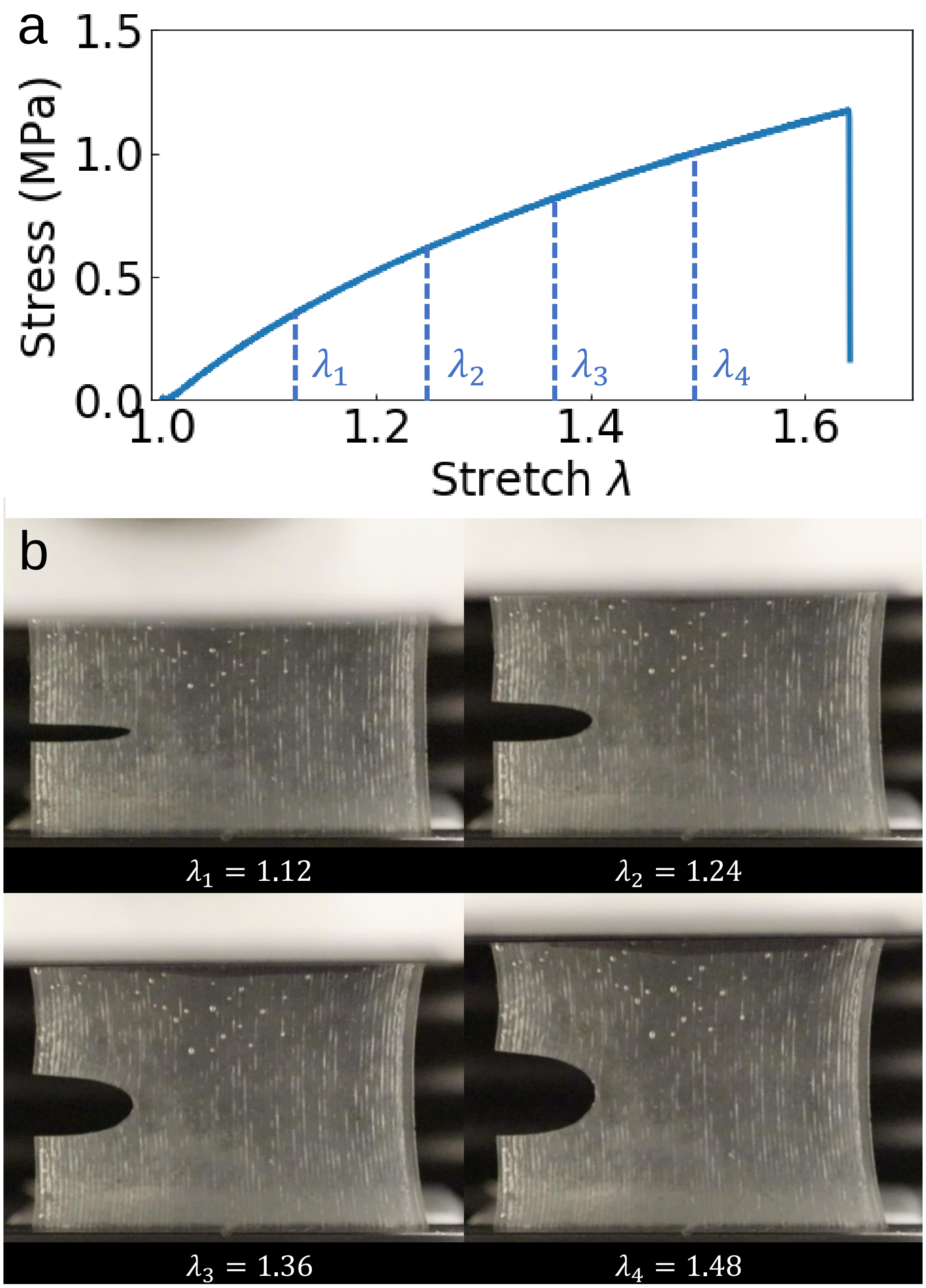

Figure S6: (a) Representative stress-stretch behavior of notched 3D printed PDUO without fibers. (b) Optical images of the deformation of the sample at indicated stretch levels.