Electronic Supplementary Material (ESI) for Nanoscale Advances.

Supplementary Information Available

Graphene Oxide Nanosheets Augment Silk Fibroin Aerogels for Enhanced Water Stability and Oil Adsorption

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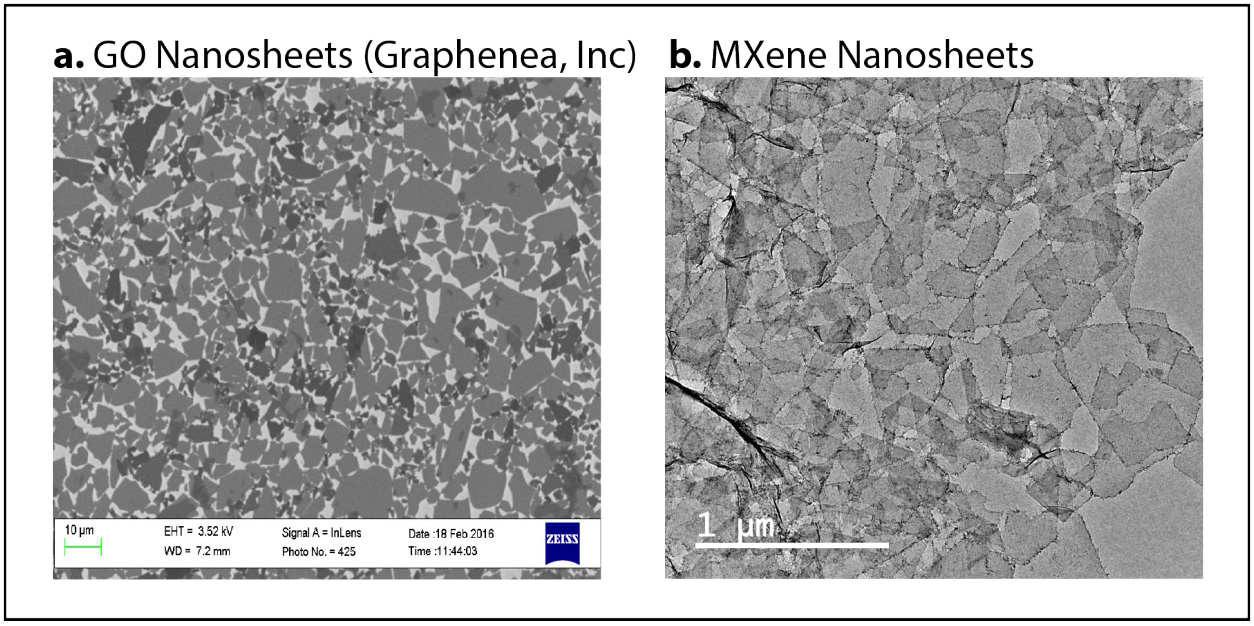

Figure S1: a) Representative SEM image of GO nanosheets (provided by Graphenea, Inc. `https://www.graphenea.com/collections/graphene-oxide/products/graphene-oxide-4-mg-ml-water-dispersion-1000-ml`). b) Representative TEM of MXene nanosheets.

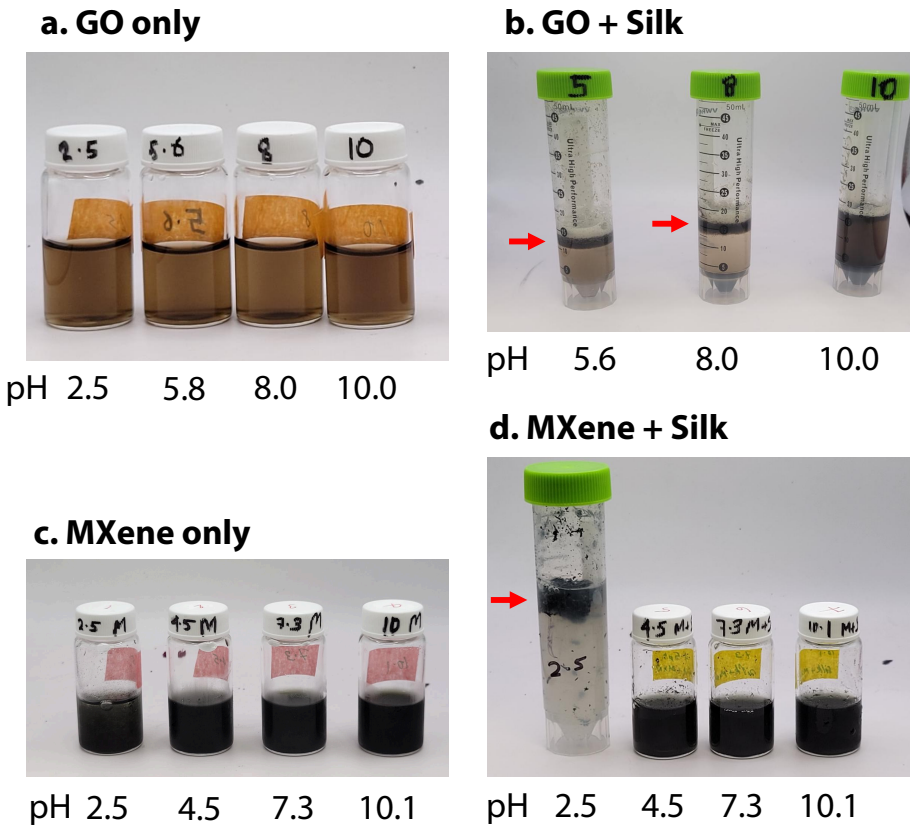

Figure S2: Dispersion of silk fibroin and 2D material nanosheets in water at varying pH, showing greater stability at pH 10. (a) 2 mg/mL graphene oxide, (b) 2 mg/mL graphene oxide with 20 mg/mL silk fibroin, (c) 2 mg/mL MXene, (d) 2 mg/mL MXene with 20 mg/mL silk fibroin. Red arrows denote aggregated conditions. (e) Zeta potential of various silk fibroin and 2D material mixtures, with N = 3 experimental replicates. Markers denote mean and error bars indicate standard deviation. Aggregated solutions at low pH were not characterized.

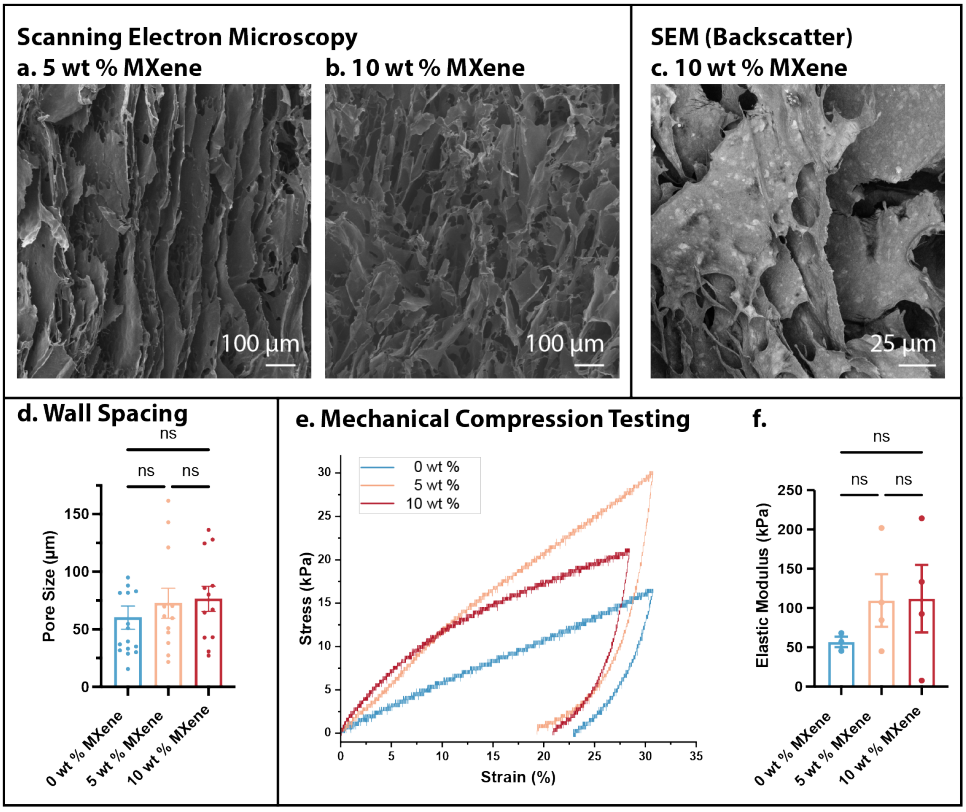

Figure S3: Microstructure and mechanical characterization of Silk-MXene aerogels. (a,b) Representative cross-sectional SEM images at 5 wt % MXene and and 10 wt % MXene. (c) Electrical conductivity of 10 wt % MXene aerogels imaged through directional backscatter mode in SEM. (d) Comparison of lateral wall separation as a readout of pore size. (e) Representative stress-strain curve of silk-MXene aerogels. (f) Elastic modulus and calculated strain recovery efficiency at 30 % strain ($N = 4$ samples tested per condition). Each point denotes a measurement on a different sample, and bar plot shows mean value with error bars denoting standard error of the mean (SEM). One-way ANOVA test for statistics * $p < 0.05$, ** $p < 0.01$

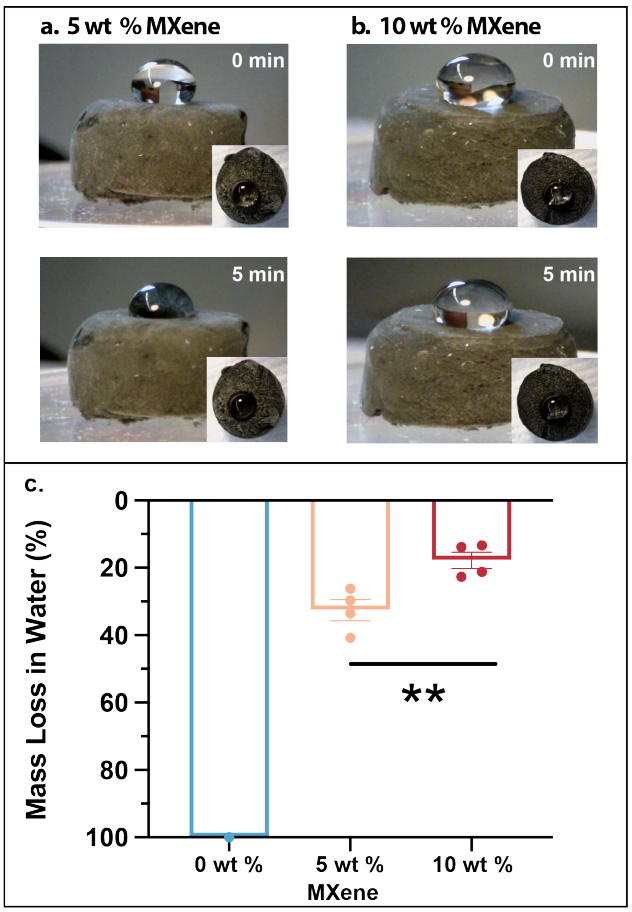

Figure S4: Water stability of MXene-silk aerogels. (a, b) Water droplet to demonstrate stability for 5 wt % and 10 wt % MXene aerogels, timed images over 5 minutes. (c) Percent of mass loss for 5 wt % and 10 wt % MXene aerogel measured after 5 days ($N = 3$ samples), Student's t-test for statistics * $p < 0.05$, ** $p < 0.01$. Silk-only aerogels were fully dissolved after 5 days in water and counted as 100% mass loss.

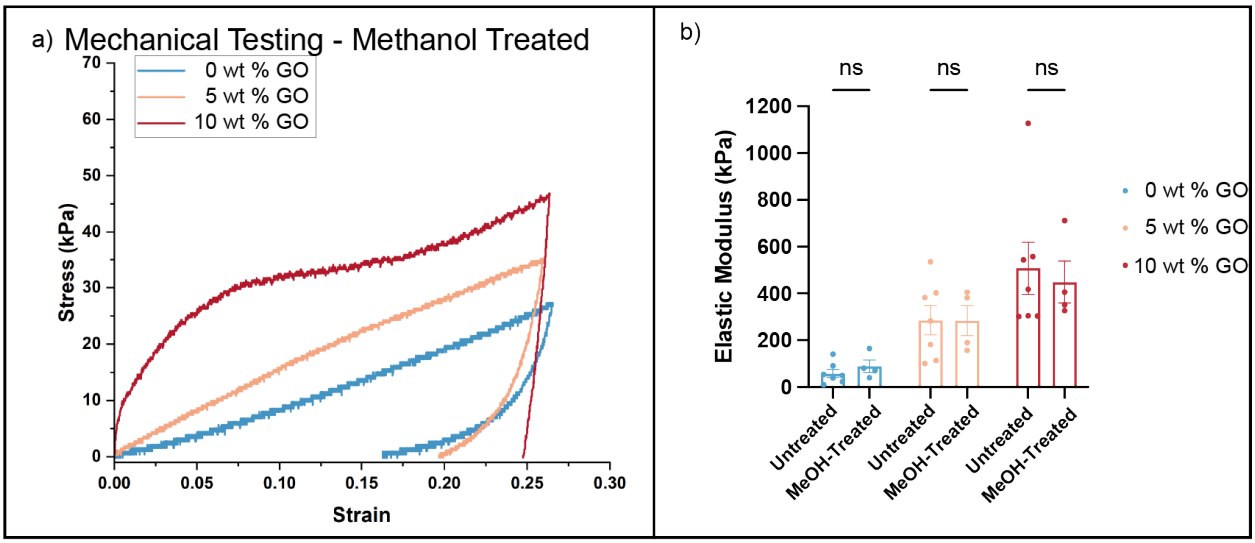

Figure S5: Mechanical testing of methanol-treated silk-GO aerogels. a) Representative stress and strain curves from compression testing on silk aerogels (0, 5, 10 wt % GO) after methanol treatment. b) Calculated elastic modulus for untreated ($N = 5$ samples) and methanol-treated aerogels (MeOH) ($N = 4$ samples). Plotted in a box-and-whisker plot as x=median value and whisker=standard error of the mean (SEM). Two-way ANOVA test for statistics * $p < 0.05$, ** $p < 0.01$.

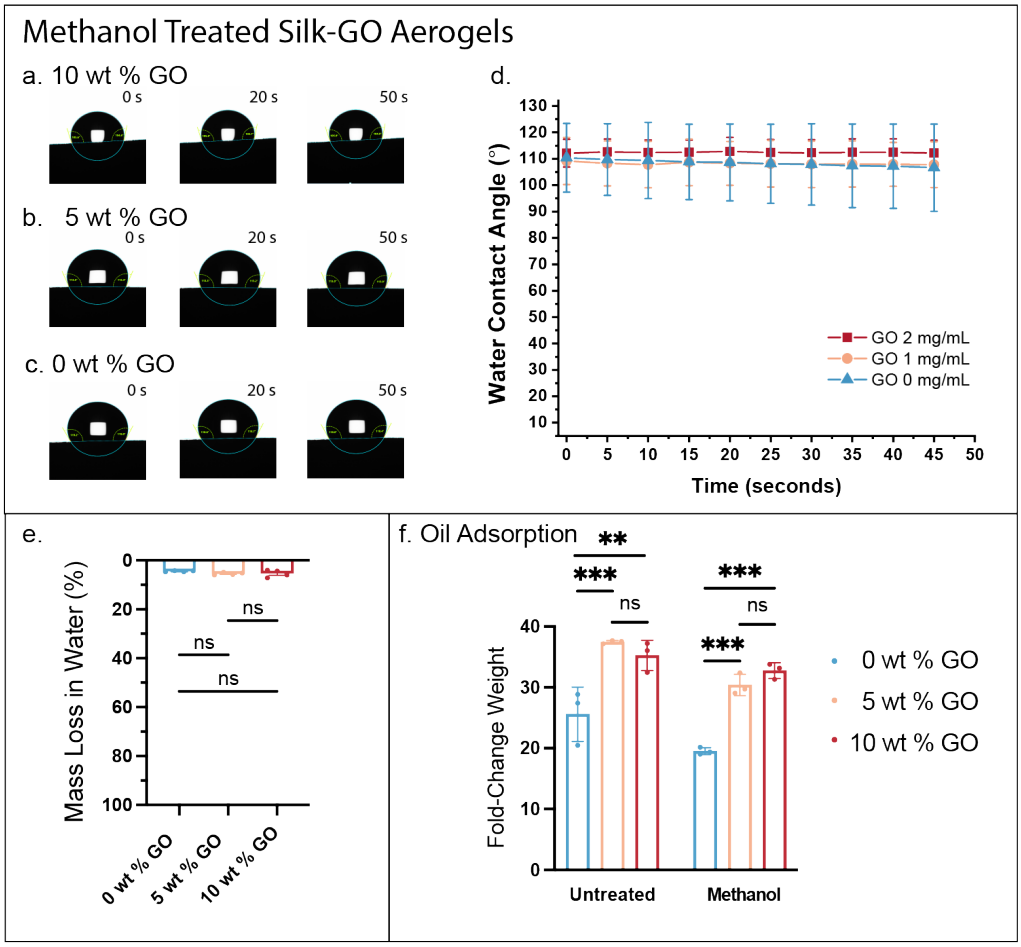

Figure S6: Interactions of methanol-treated silk-GO aerogels with water. (a-c) Representative water contact angle for silk aerogels (10, 5, 0 wt % GO). (d) Average contact angle of water after contacting aerogel surface ($N = 3$ samples). (e) Percentage of mass loss for all concentrations for methanol treated aerogels, plotted in a bar plot as x=median value and whisker=standard error of the mean (SEM) ($N = 4$ samples), One-way ANOVA test. (f) Measured napthenic oil absorption for methanol treated aerogels (fold change by weight), plotted in a bar plot as x=mean value and error bars are standard error of the mean (SEM) (N=4). Two-way ANOVA test, * $p < 0.05$, ** $p < 0.01$.

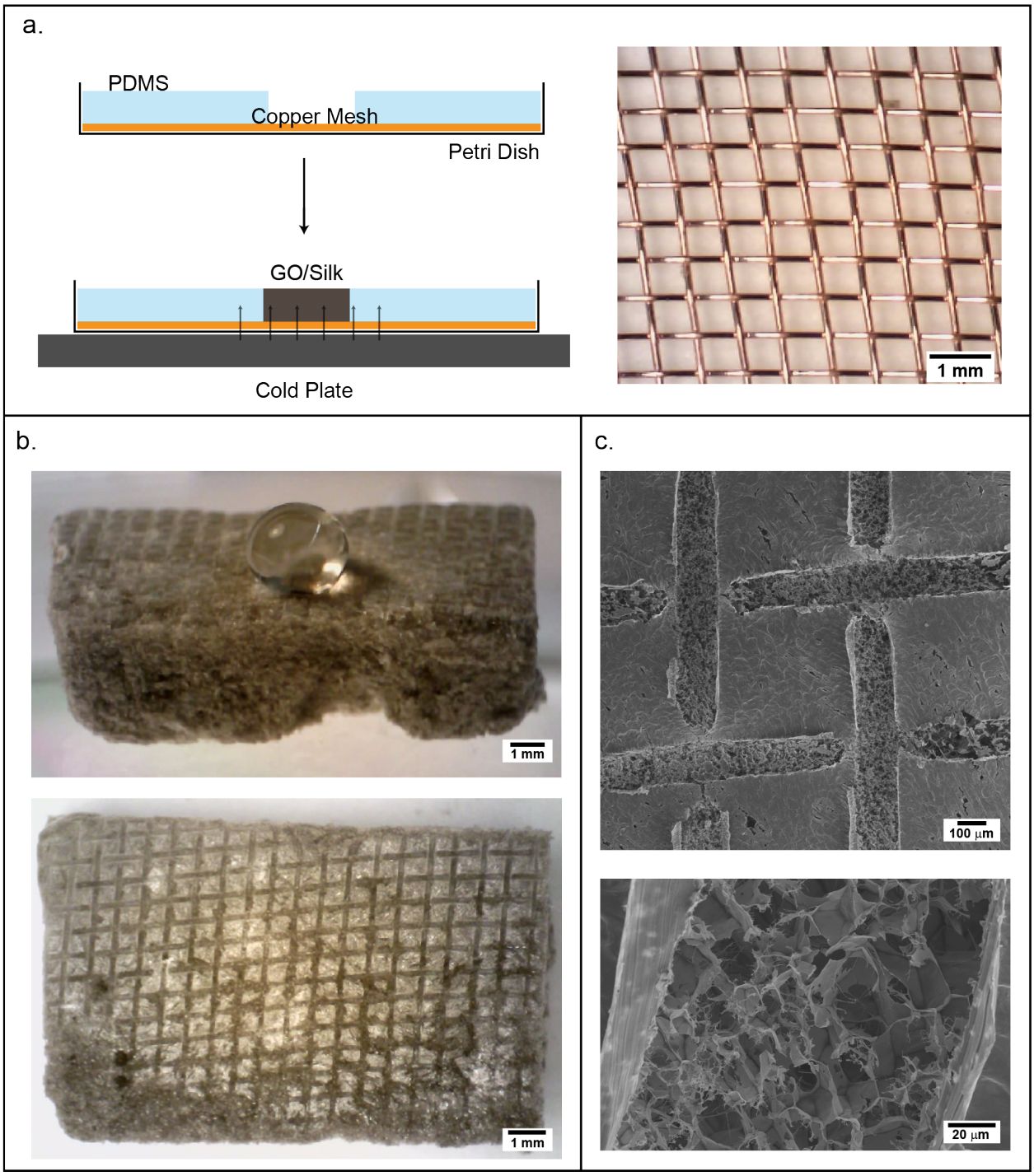

Figure S7: Large area deposition of silk-GO aerogels. a) Schematic of experimetnal setup and representative image of pristine copper mesh. (b) Cross-section and top-down view of silk aerogel with 10 wt% GO after casting on a copper mesh. Water droplet added in cross sectional view to illustrate hydrophobic properties. (c) SEM of aerogel surface after casting on copper mesh.