Electronic Supplementary Material (ESI) for Soft Matter.

Tunable Hygromorphism via Low Molecular Weight Gel Reinforced – Electrospun Mat Composites

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SUPPLEMENTAL INFORMATION

Supplemental Videos:

Preferential Curvature of Controls and Bilayers:

Supplemental Video 1: **Control Layer A1.mp4**

Description: Preferential curvature of active layer A1 set in a helical conformation in water

Supplemental Video 2: **Control Layer A2.mp4**

Description: Preferential curvature of active layer A2 set in a helical conformation in water

Supplemental Video 3: **Bilayer A1-P1_towards mat coil.mp4**

Description: Preferential curvature of bilayer A1-P1 set in a "towards mat" helical conformation in water

Supplemental Video 4: **Bilayer A2-P1_towards mat coil.mp4**

Description: Preferential curvature of bilayer A2-P1 set in a "towards mat" helical conformation in water

Supplemental Video 5: **Bilayer A1-P1_away from mat coil.mp4**

Description: Preferential curvature of bilayer A1-P1 set in an "away from mat" helical conformation in water

Supplemental Video 6: **Bilayer A2-P1_away from mat coil.mp4**

Description: Preferential curvature of bilayer A2-P1 set in an "away from mat" conformation in water

Natural Curvature of Controls and Bilayers

Supplemental Video 7: **A1 control.mp4**

Description: Natural curvature of active layer A1 in water

Supplemental Video 8: **A2 control.mp4**

Description: Natural curvature of active layer A2 in water

Supplemental Video 9: **A1-P1 bilayer.mp4**

Description: Natural curvature of bilayer A1-P1 in water

Supplemental Video 10: **A2-P1 bilayer.mp4**

Description: Natural curvature of bilayer A2-P1 in water

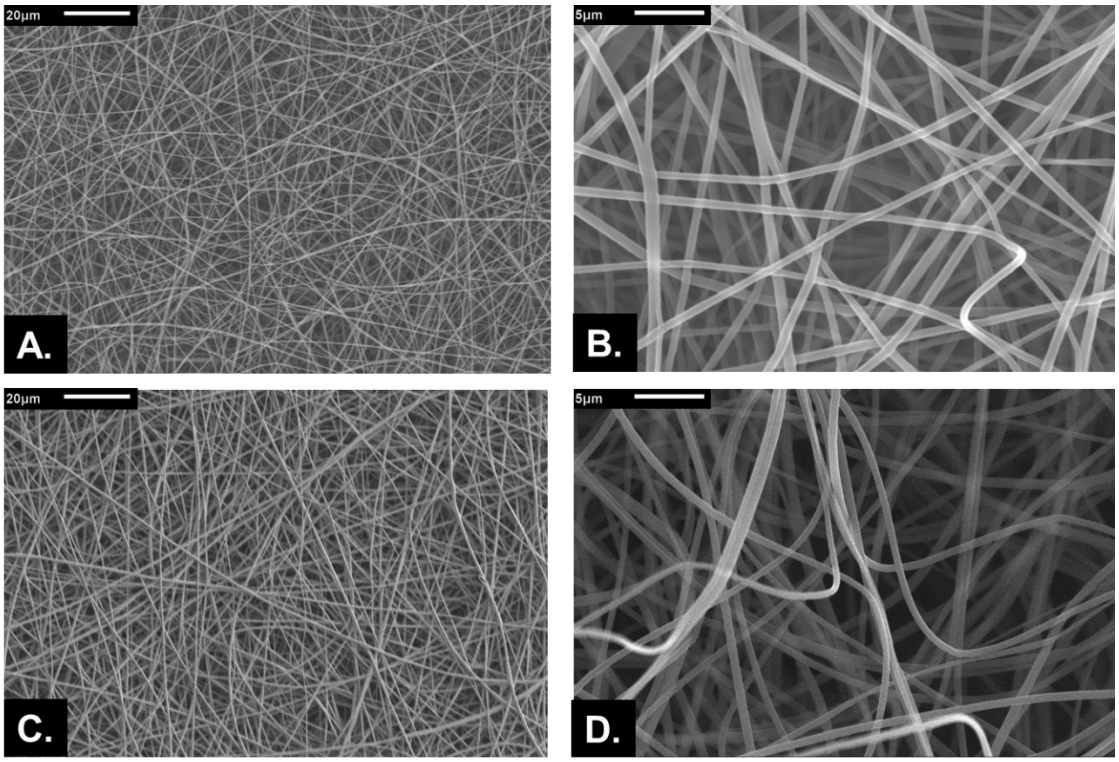


Figure S1: Scanning electron microscopy (SEM) of PVA nanofiber mats

A. & B. PVA electrospun nanofiber mat used in control layer A1 (Fiber Size = 0.337 ± 0.07 µm; Mat thickness ~500 µm)

C. & D. PVA electrospun nanofiber mat used in control layer A2 (Fiber Size = 0.328 ± 0.05 µm; Mat thickness ~1000 µm)

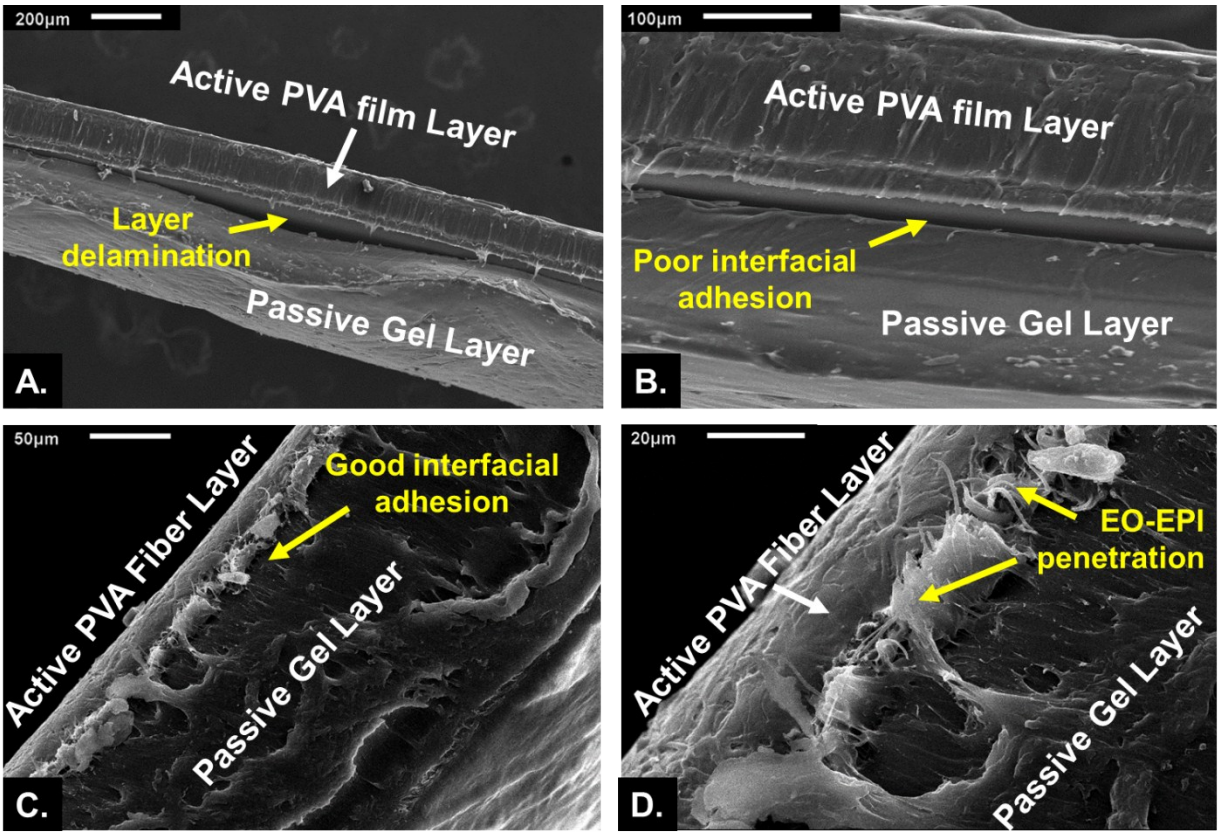
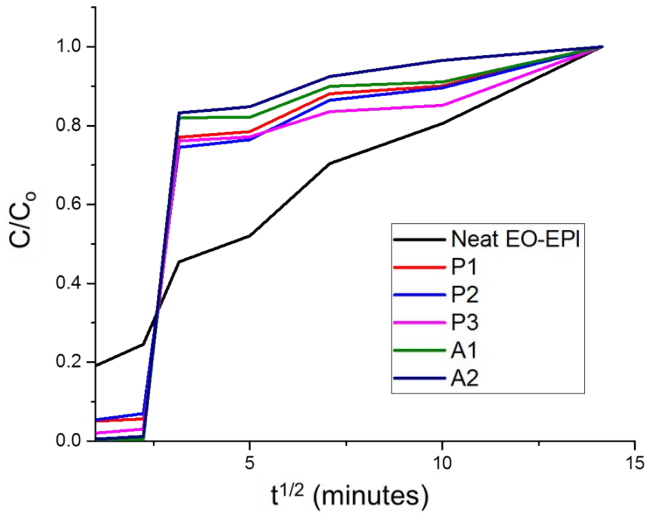


Figure S2: Cross-sections of bilayer composites containing either a PVA film or nanofiber mat

A. & B. SEM of PVA film bilayer composite cross-section highlighting delamination and poor interfacial adhesion.

C. & D. SEM of PVA nanofiber bilayer composite cross-section highlighting penetration of the EO-EPI coat layer through the fibers and good interfacial adhesion



Sample	Thickness, h (m)	Diffusion Coefficient, D (m^2 min^{-1})
Neat EO-EPI	0.00014	**1.56E-11**
P1 Control Layer	0.00009	**1.14E-11**
P2 Control Layer	0.0001	**1.21E-11**
P3 Control Layer	0.0001	**1.43E-11**
A1 Control Layer	0.00015	**1.98E-11**
A2 Control Layer	0.0002	**3.30E-11**

Figure S3: Control layer diffusion profiles and calculated diffusion coefficients

Based on a calibration curve, the concentration of dye in each aliquot was determined using UV/Vis spectroscopy. The following equation describes the change in dye concentration with time:

$$\left(\frac{C}{C_o}\right) = \sqrt{\frac{D}{h^2\pi}}t^{\frac{1}{2}}$$

where C is the concentration of dye at time t, C_o is the total concentration of dye in the original sample, D is the diffusion coefficient, h is the sample thickness, and t is time in minutes. C/C_o vs. $t^{1/2}$ was plotted to determine the diffusion coefficient from the slope. Due to the initial burst release of the control layers caused by surface dye elution, the slope was determined starting at $t^{1/2} = 5^{1/2}$. The values for C and C_o as a function of time were obtained using the concentrations determined by UV/Vis spectroscopy.

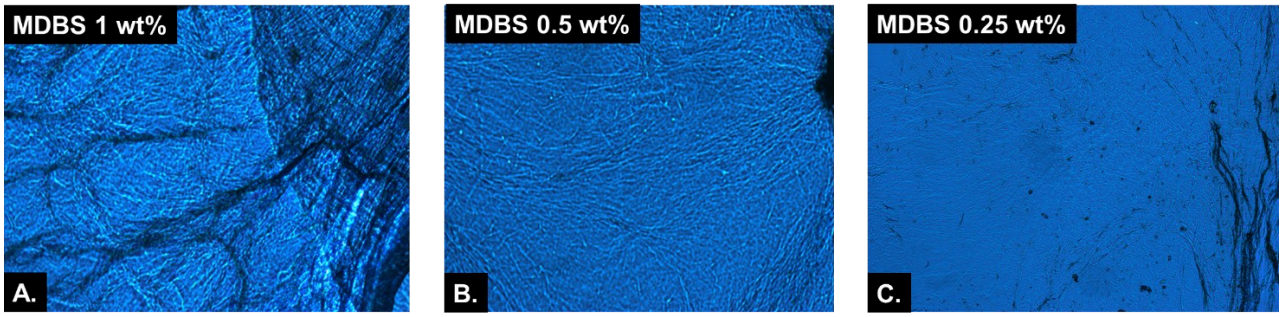


Figure S4: Polarized light microscopy of MDBS gels at different weight fractions

A. MDBS 1 wt% gel used for control passive layer **P1**; **B.** MDBS 0.5 wt% gel used for control passive layer **P2**; **C.** MDBS 0.25 wt% used for control passive layer **P3**

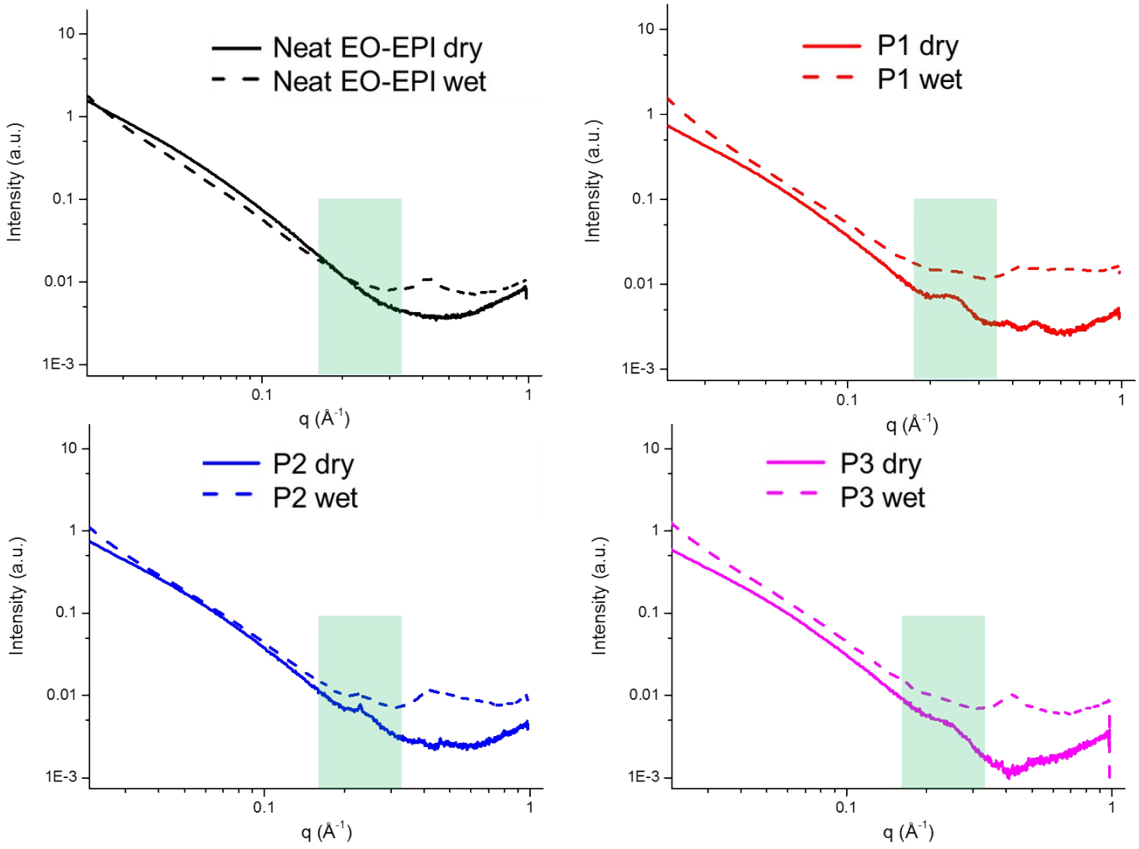


Figure S5: Small-angle X-ray Scattering (SAXS) of neat EO-EPI and wet and dry passive layer controls (**P1- P3**).

The presence of the MDBS network peak between q values 0.2 and 0.3 inverse angstroms in both wet and dry composite films confirms the preservation of the MDBS nanofiber assembly. This scattering behavior correlates with the similar wet and dry moduli for P1 and P2**.** The peak that evolves from wet EO-EPI (q = 0.4 $Å^{-1}$) is due to the exclusion of the hydrophobic epichlorohydrin (EPI) segment from the water phase surrounding the hydrophilic ethylene oxide (EO) segment.