

**Programming Shape and Tailoring Transport: Advancing Hygromorphic Bilayers with  
Aligned Nanofibers**

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SUPPLEMENTAL INFORMATION

## **Supplemental Videos:**

### ***Hygromorphic Curvature of Bilayers:***

*Supplemental Video 1 (SV1):*

**Description:** Natural curvature of bilayer composite with vertically-aligned PVA nanofibers

*Supplemental Video 2 (SV2):*

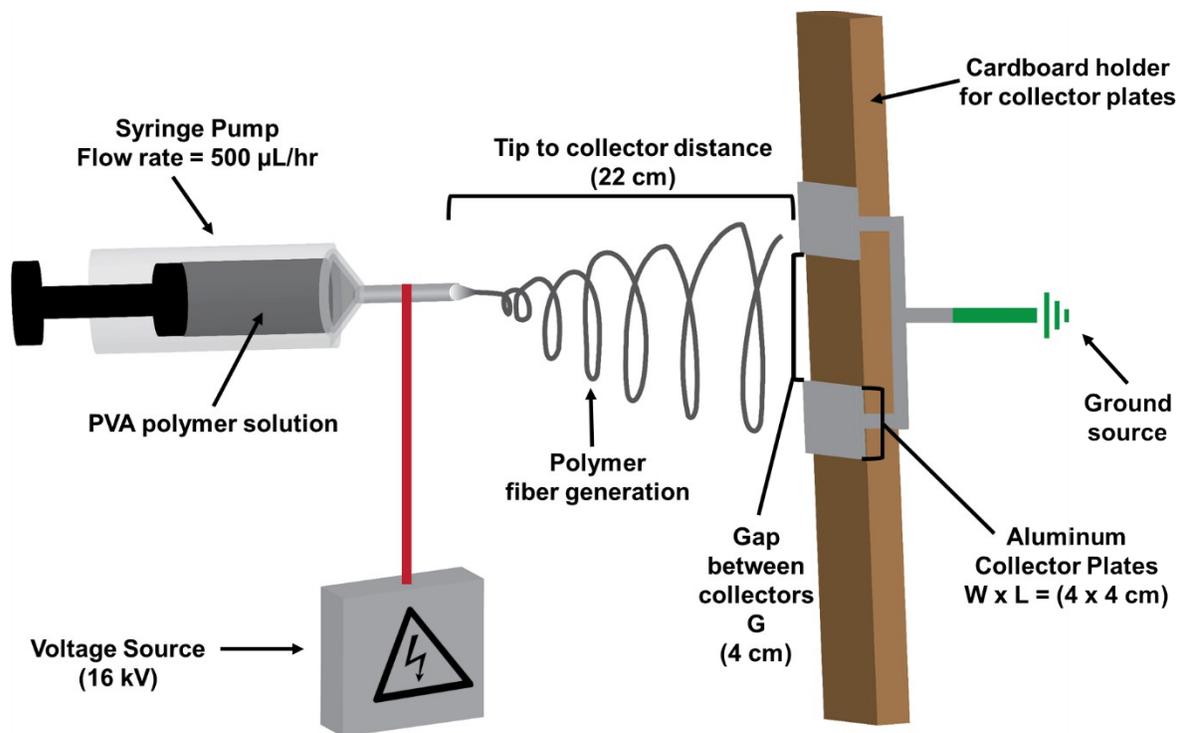
**Description:** Natural curvature of bilayer composite with horizontally-aligned PVA nanofibers

*Supplemental Video 3 (SV3):*

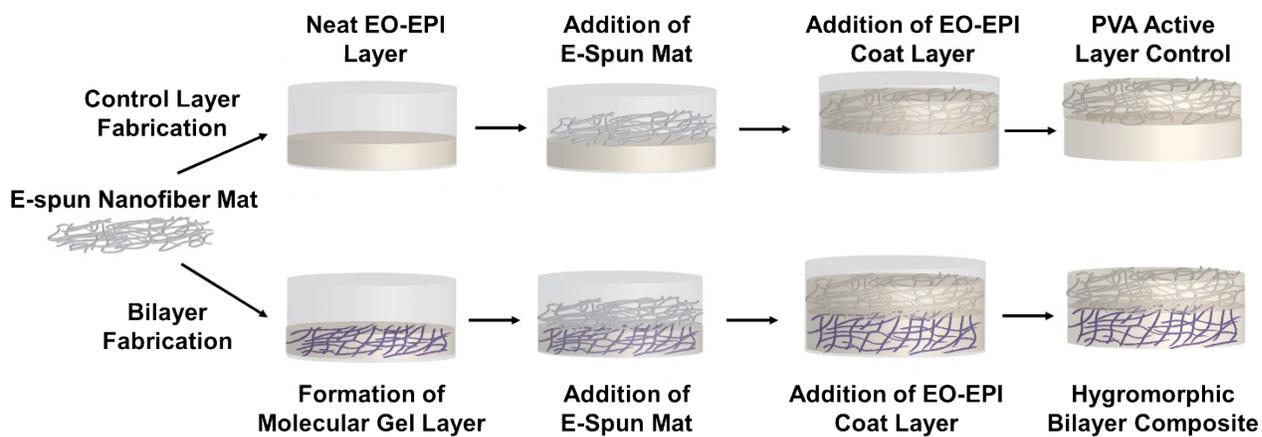
**Description:** Natural curvature of bilayer composite with diagonally-aligned PVA nanofibers

*Supplemental Video 4 (SV4):*

**Description:** Lifting of weight by horizontally-aligned composite



**Figure S1:** Schematic of parallel plate collector used to obtain aligned nanofibers



**Figure S2:** Schematic of Control Active Layers vs. Bilayer Composites

**Table S1:** Calculated Diffusion Coefficients for Random and Aligned Nanofiber Active Layers

<b>Sample</b>	<b>Thickness, h (m)</b>	<b>Diffusion Coefficient, D (m<sup>2</sup> min<sup>-1</sup>)</b>
<b>EO-EPI + Random PVA Nanofibers</b>	0.00014	<b>1.56E-11</b>
<b>EO-EPI + 0° Aligned PVA Nanofibers</b>	0.00011	<b>1.26E-10</b>
<b>EO-EPI + 90° Aligned PVA Nanofibers</b>	0.00011	<b>2.10E-10</b>
<b>EO-EPI + 45° Aligned PVA Nanofibers</b>	0.00011	<b>4.09E-11</b>

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:

$$\left(\frac{C}{C_0}\right) = \sqrt{\frac{D}{h^2\pi}} t^{\frac{1}{2}}$$

where C is the concentration of dye at time t, C<sub>0</sub> 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<sub>0</sub> vs. t<sup>1/2</sup> 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<sup>1/2</sup> = 5<sup>1/2</sup>. The values for C and C<sub>0</sub> as a function of time were obtained using the concentrations determined UV/Vis spectroscopy.

**Table S2:** Layer Properties for Electrospun Nanofiber Mats and Corresponding Control Layers

<b>Sample</b>	<b>E-spun Mat Crystallinity (%)</b>	<b>E-spun Mat Fiber Size (nm)</b>	<b>Layer Thickness (mm)</b>	<b>Layer Expansion Coefficient, <math>\alpha</math></b>	<b>Layer Young's Modulus, Hydrated (MPa)</b>
<b>Neat EO-EPI</b>	N/A	N/A	0.14 mm	0.6	$0.17 \pm 0.04$
<b>EO-EPI + Random PVA Nanofibers</b>	$63 \pm 4$	$571 \pm 37$	$0.12 \pm 0.04$	1.2	$0.12 \pm 0.04$
<b>EO-EPI + 0° Aligned PVA Nanofibers</b>	$64 \pm 2$	$534 \pm 24$	$0.11 \pm 0.05$	1.4	$0.081 \pm 0.006$
<b>EO-EPI + 90° Aligned PVA Nanofibers</b>	$64 \pm 2$	$534 \pm 24$	$0.11 \pm 0.05$	1.4	$0.076 \pm 0.001$
<b>EO-EPI + 45° Aligned PVA Nanofibers</b>	$64 \pm 2$	$534 \pm 24$	$0.11 \pm 0.05$	1.5	$0.054 \pm 0.011$