

Supporting Information for:

# Reinventing Butyl Rubber for Stretchable Electronics

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## Additional Experimental Information

$\text{Ru}(\text{dtb-bpy})_3(\text{PF}_6)_2$  and  $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$  were synthesized according to previously published methods.<sup>[1,2]</sup> Silver nanowire (AgNW) stock dispersions (10 mg/mL in ethanol) were purchased from Blue Nano Inc. Indium tin oxide (ITO) on glass (15-25  $\Omega/\text{sq}$ ) (Delta Technologies). Patterned indium tin oxide (ITO) was obtained from Thin Film Devices, Inc., CA. Conductive copper tape was obtained from Ted Pella, Inc., CA. AFM images were processed using WSXM 5.0 Develop 8.0 software.<sup>[3]</sup>

*Preparation of Fluorinated Glass Molding Masters:* We deposited SAMs of trichloro(1H,1H,2H,2H-perfluorooctyl)silane on UV ozone (Jelight UV Ozone cleaner, Model No. 42A) cleaned glass plates using vapor deposition for 20 h in a vacuum desiccator.

### Barrier Studies

*Organic-Inorganic Perovskite ( $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$ ) Films:* 0.500 g  $\text{CH}_3\text{NH}_3\text{I}$  and 0.292 g lead (II) chloride ( $\text{PbCl}_2$ ) were mixed in 3 mL anhydrous N,N-dimethylformamide (DMF) and heated at 60 °C for 24 h with stirring to prepare the mixed-halide perovskite ( $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$ ) solution. The  $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$  solution was then spin-coated on UV ozone cleaned glass substrates at 1500 rpm for 45 s. PDMS and T-IIR barriers (0.5 mm thick) were then laminated on top of the films. The samples were treated with 100% RH at 75 °C in a Cincinnati Sub-Zero humidity and temperature control chamber. Absorption spectra were measured using a Varian Cary 50 UV-Visible Spectrophotometer. The T-IIR barrier was removed prior to spectrum acquisition to avoid the contribution of the T-IIR absorption to the spectrum (Figure S2).

*Light-Emitting Electrochemical Cells:* Patterned ITO glass substrates were sonicated for 15 min in deionized  $\text{H}_2\text{O}$  and 15 min in isopropanol followed by treatment with UV ozone for 5 min. The  $\text{Ru}(\text{dtb-bpy})_3(\text{PF}_6)_2/\text{PMMA}$  emissive layer (3:1 v/v mixture of a 40 mg/mL solution of  $\text{Ru}(\text{dtb-bpy})_3(\text{PF}_6)_2$  in acetonitrile and a 25 mg/mL solution of PMMA in acetonitrile) was then spin-coated (1000 rpm for 30

s) on patterned ITO in a glove box. The edges of the patterned ITO were swabbed using acetonitrile to expose parts of the ITO to be used as electrodes in the device. The resulting film was then annealed in an oven overnight at 120 °C. 500 Å gold electrodes were then directly deposited on the Ru(dtb-bpy)<sub>3</sub>(PF<sub>6</sub>)<sub>2</sub>/PMMA emissive layer through a stainless steel shadow mask by electron beam evaporation. The rate of evaporation was increased stepwise from 0.5-1.0 Å/s. PDMS and T-IIR barriers were then laminated on top of the LEECs, and the edges of the barrier layers were sealed using polyimide tape. The Au and ITO pattern served as six individual devices on a single glass substrate (Figure S3). Three out of six devices on each glass substrate were characterized in an inert atmosphere (glovebox) prior to humidity treatment. The remaining three devices were characterized after exposure to 100% RH in the humidity controlled chamber for 3 h at 25 °C. LEEC devices were characterized using a voltage of 5 V DC. Radiance was measured using an integrating sphere and a calibrated UDT S470 optometer. Current was measured using a Keithley 2601A SourceMeter.

*Copper and Silver Films:* Glass substrates (2.5 cm x 2.5 cm) were cleaned by sonication in deionized H<sub>2</sub>O and isopropanol for 15 min each in a sonicator followed by treatment with UV ozone for 5 min. 15 Å of titanium as an adhesion layer followed by 250 Å of copper or silver was deposited in an e-beam evaporator at a deposition rate of 2 Å/s onto the glass substrate through a shadow mask comprising 1 mm x 10 mm openings. PDMS and T-IIR barriers were then laminated on top of the copper and silver thin-film wires, and the edges of the barrier layers were sealed using polyimide tape. The LED circuits were fabricated by depositing two 250 Å thick Ag wires (2 mm x 20 mm) on glass, and then connecting these thin-film wires in series with a surface mount LED (Lumex Standard LED, 1206) using conductive adhesive (Effective Shielding, PA). The LED circuits were laminated using PDMS and T-IIR barrier layers and operated with a current source of 1mA. The encapsulated wires and LED circuits were exposed to 100% RH in a humidity control chamber. For underwater testing, LED circuits were

fabricated by depositing four 250-Å-thick Ag wires (2 mm x 20 mm) on glass and then connecting these thin-film wires in series with two surface mount LEDs and a 250 Ω surface mount resistor (KOA Speer). - PDMS and T-IIR barrier layers were then laminated on top of the LED circuits, and the edges of the barrier layers were sealed using polyimide tape. The LED circuits were then immersed in water in a beaker, and operated at 5 V DC. Optical inspection was carried out using an Olympus BX51M optical microscope. Optical micrographs were captured using an Olympus Qcolor 3 digital microscope camera. Resistances of encapsulated wires and voltage drop across encapsulated LED circuits were recorded using Keithley Sourcemeter.

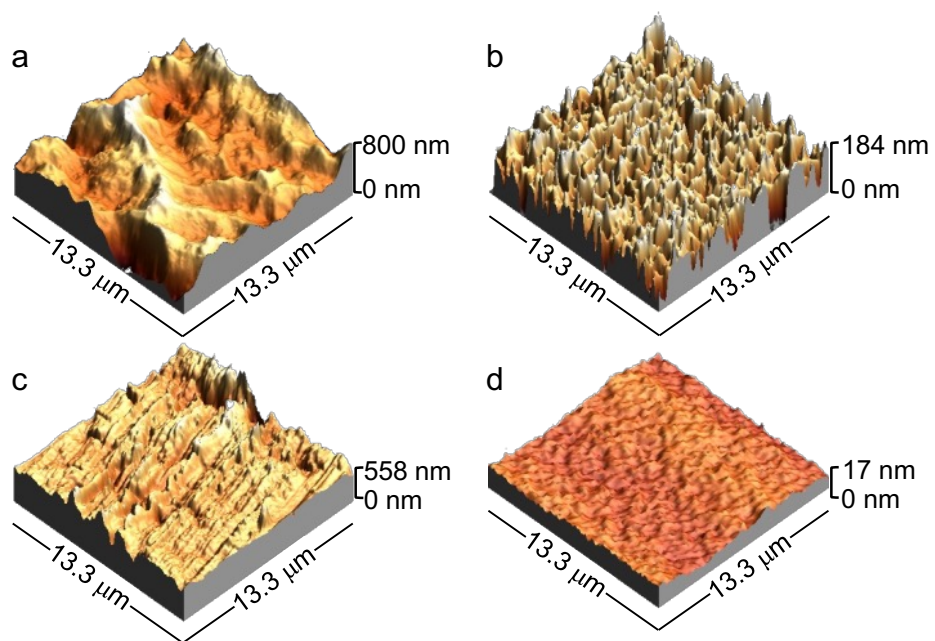
*Silver Nanowire (AgNW) Films:* Glass slides were cleaned by sonication in deionized H<sub>2</sub>O and isopropanol for 15 min each in a sonicator followed by treatment with air plasma for 1 min at an air pressure of 10 psig and a flow rate of 9.2 mL/min at medium discharge setting. T-IIR substrates were sonicated in acetone and isopropanol for 10 min each, and then treated with oxygen plasma for 15 min at O<sub>2</sub> pressure of 10 psig and flow rate of 10.6 mL/min at medium discharge setting. The oxidized T-IIR substrates were swabbed with isopropanol and dried in a stream of nitrogen. The AgNW dispersion was diluted in anhydrous ethanol to 0.6 mg/mL and sonicated for 20 s to reduce AgNW aggregation. The dispersion (0.3 mL) was then drop cast onto the oxidized glass surfaces and T-IIR<sub>[ox]</sub> surfaces. The samples were allowed to dry on a KS 130 basic (IKA) shaker at 160 rpm at room temperature. A 0.5-cm-wide border was swabbed with isopropanol to remove nanowire aggregates around the edge of each sample, and then dried under a stream of nitrogen. PDMS and T-IIR barriers were laminated onto the AgNW films on glass, and T-IIR barriers were laminated onto the AgNW films on T-IIR<sub>[ox]</sub>. The barrier layers were allowed to wet the 0.5-cm-wide border of the substrate surrounding the film, forming a seal around it. The laminated AgNW samples were placed in a vapor chamber along with unprotected AgNW reference samples. 10 drops of HNO<sub>3</sub> were added to the chamber, and the samples were exposed

to HNO<sub>3</sub> vapor for 12 h, removed from the chamber, and the barrier layers were then carefully removed from the samples. Optical inspection was carried out using an Olympus BX51M optical microscope with dark field illumination. Optical micrographs were captured using an Olympus Qcolor 3 digital microscope camera. Sheet resistance measurements were carried out using a four-point wire setup. A minimum of three measurements from three samples were averaged. Absorption spectra were measured using a Varian Cary 50 UV-Visible Spectrophotometer.

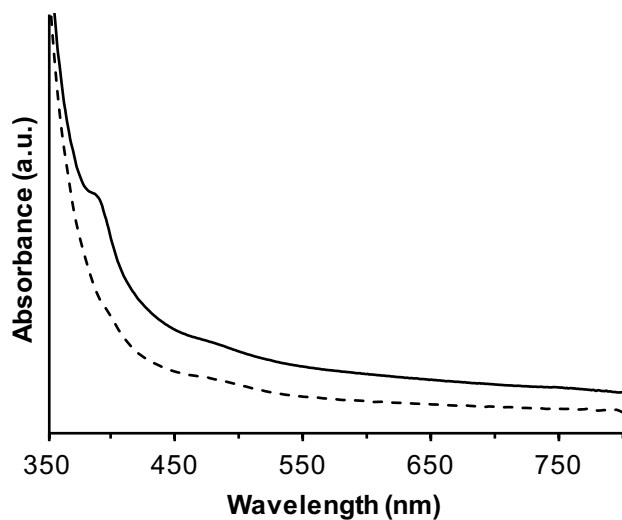
**Table S1.** Comparison of mechanical properties of T-IIR and PDMS.

Physical property	T-IIR	PDMS <sup>[a]</sup>
Hardness (Shore A)	40	48.3
Tensile strength (MPa)	3.51	6.25
Ultimate elongation (%)	170	120
Young's Modulus @ <40 % strain (MPa)	0.41	2.05

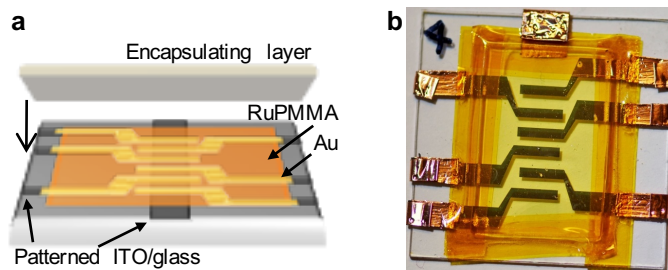
<sup>[a]</sup> Values for PDMS cured at 100 °C taken from reference [4].



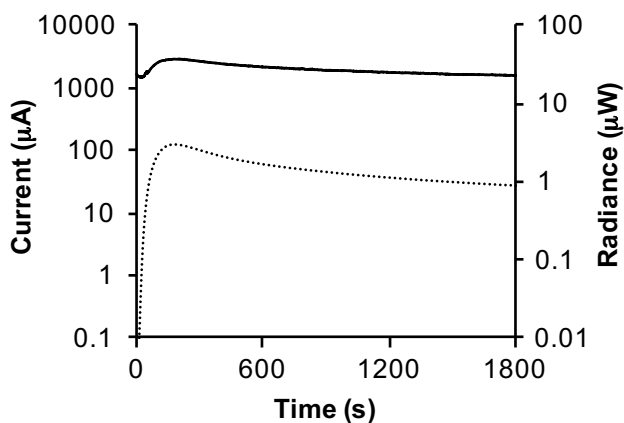
**Figure S1.** AFM topographic images of (a) T-IIR molded between Teflon sheets; (b) T-IIR molded between glass substrates; (c) Al sheet and (d) PDMS-coated Al sheet.



**Figure S2.** UV-vis absorption spectrum of T-IIR (dashed line) and T-IIR/perovskite/glass (solid line).



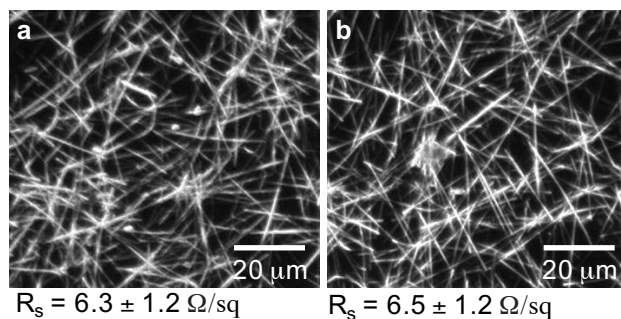
**Figure S3.** (a) Diagram of LEEC devices used for barrier studies. (b) Photograph of a T-IIR-laminated LEEC.



**Figure S4.** Evolution of current (solid line) and radiance (dotted line) response over a 30-minute testing period for an unencapsulated LEEC characterized under inert conditions.

**Table S2.** Characteristics of LEECs with no encapsulating layer characterized under inert conditions.

Average radiance ( $\mu\text{W}$ )	$2.64 \pm 0.31$
Average current (mA)	$1.52 \pm 0.28$
Average maximum EQE (%)	$0.03 \pm 0.003$



**Figure S5.** Optical microscope images of AgNW films sandwiched between T-IIR (a) before and (b) after 12 hours of nitric acid vapor exposure.

### References:

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