

## **Electrospinning-Induced Elastomeric Properties of Conjugated Polymers for Extremely Stretchable Nanofibers and Rubbery Optoelectronics**

Jung-Yao Chen,<sup>a,\*</sup> Hui-Ching Hsieh,<sup>b</sup> Yu-Cheng Chiu,<sup>c</sup> Wen-Ya Lee,<sup>d</sup> Chih-Chien Hung,<sup>e</sup> Chu-Chen Chueh<sup>b,f</sup> and Wen-Chang Chen<sup>b,e,f\*</sup>

<sup>a</sup> Department of Chemical Engineering

National Chung Cheng University

Chiayi 62102, Taiwan

E-mail: [chmjyc@ccu.edu.tw](mailto:chmjyc@ccu.edu.tw)

<sup>b</sup> Department of Chemical Engineering

National Taiwan University

Taipei 10617, Taiwan

E-mail: [chenwc@ntu.edu.tw](mailto:chenwc@ntu.edu.tw)

<sup>c</sup> Department of Chemical Engineering

National Taiwan University of Science and Technology

Taipei 10607, Taiwan

<sup>d</sup> Department of Chemical Engineering & Biotechnology and Research and Development Center for

Smart Textile Technology

National Taipei University of Technology

Taipei 10608, Taiwan

<sup>e</sup> Institute of Polymer Science and Engineering,

National Taiwan University

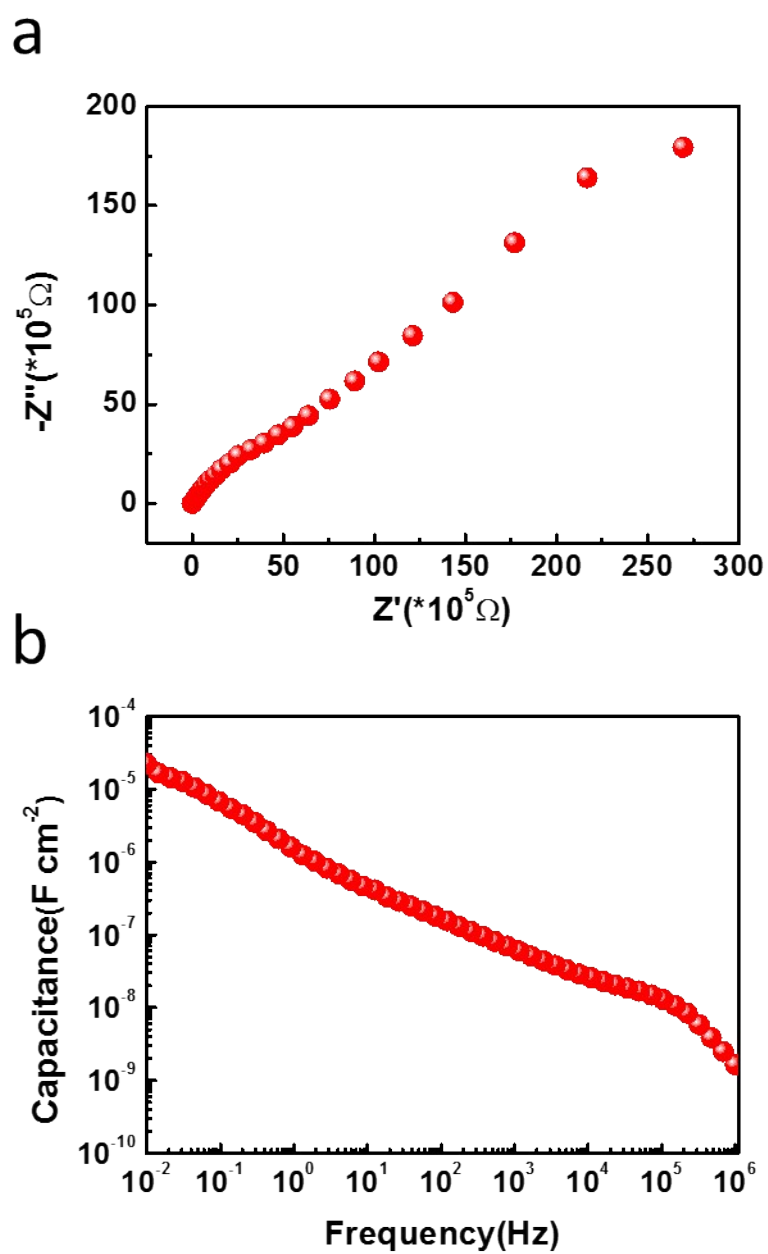
Taipei 10617, Taiwan

E-mail: [chenwc@ntu.edu.tw](mailto:chenwc@ntu.edu.tw)

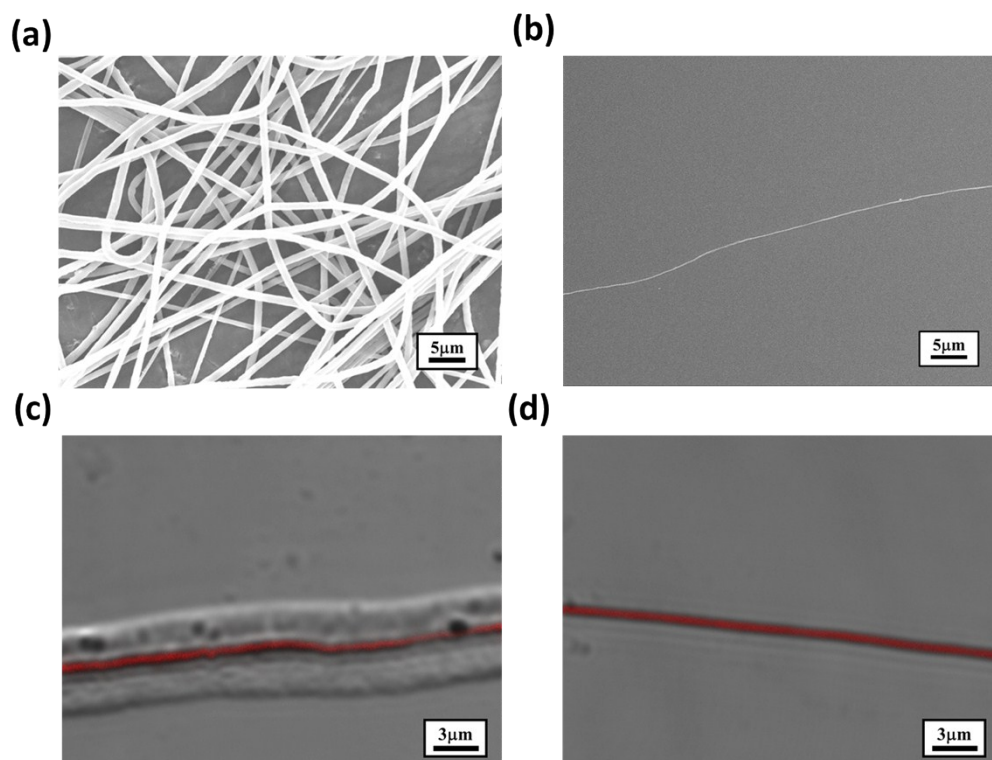
<sup>f</sup> Advanced Research Center for Green Materials Science and Technology, National Taiwan University,

Taipei 10617, Taiwan

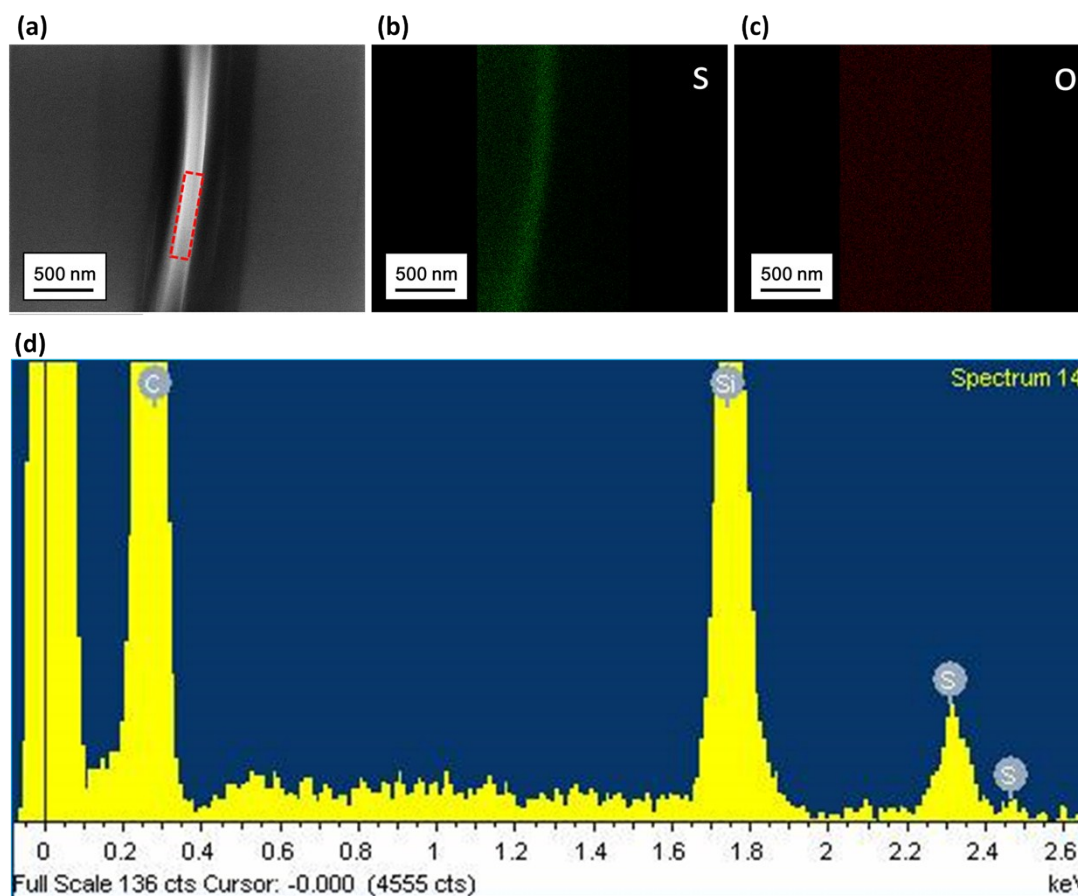
E-mail: [chenwc@ntu.edu.tw](mailto:chenwc@ntu.edu.tw)



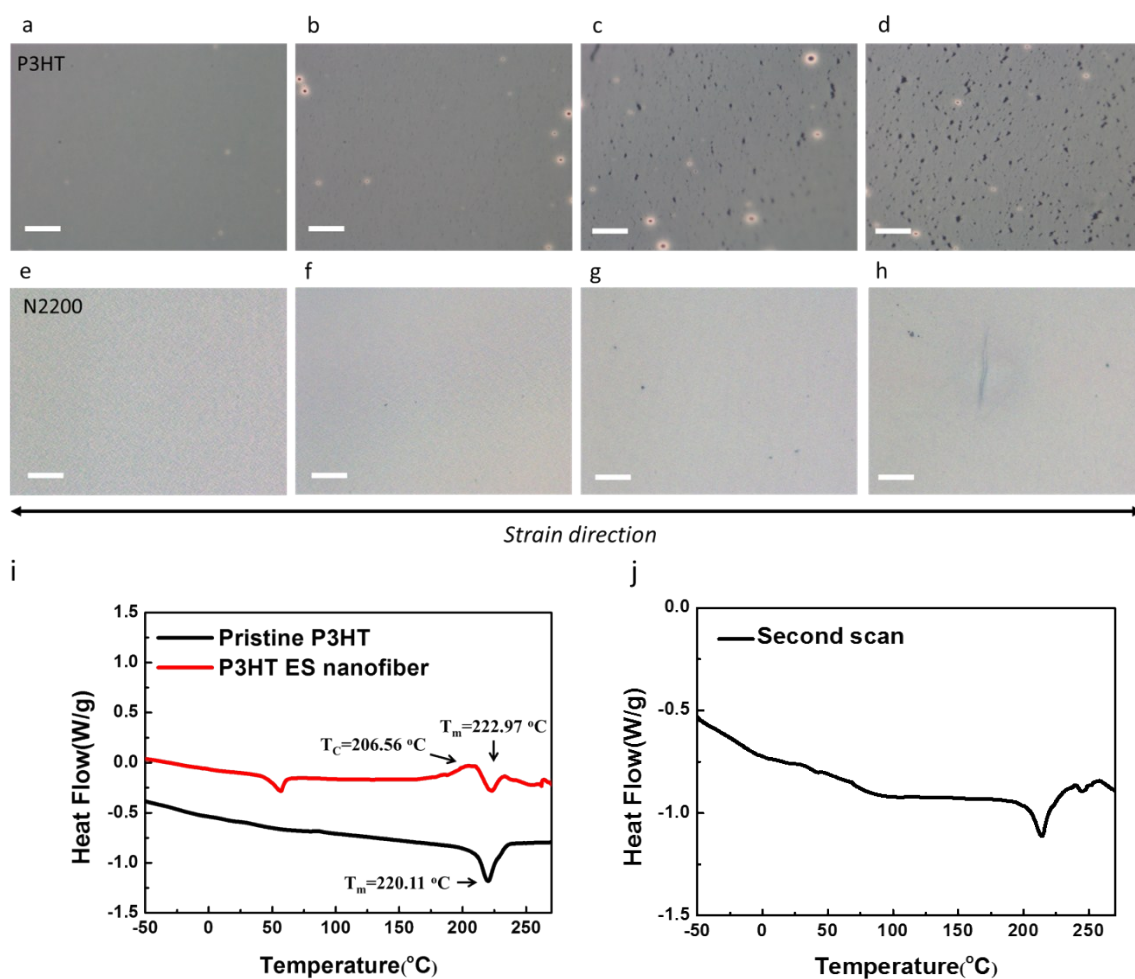
**Fig. S1** (a) The Nyquist plot and (b) the capacitance under various frequencies of the studied ion gel-based capacitor.



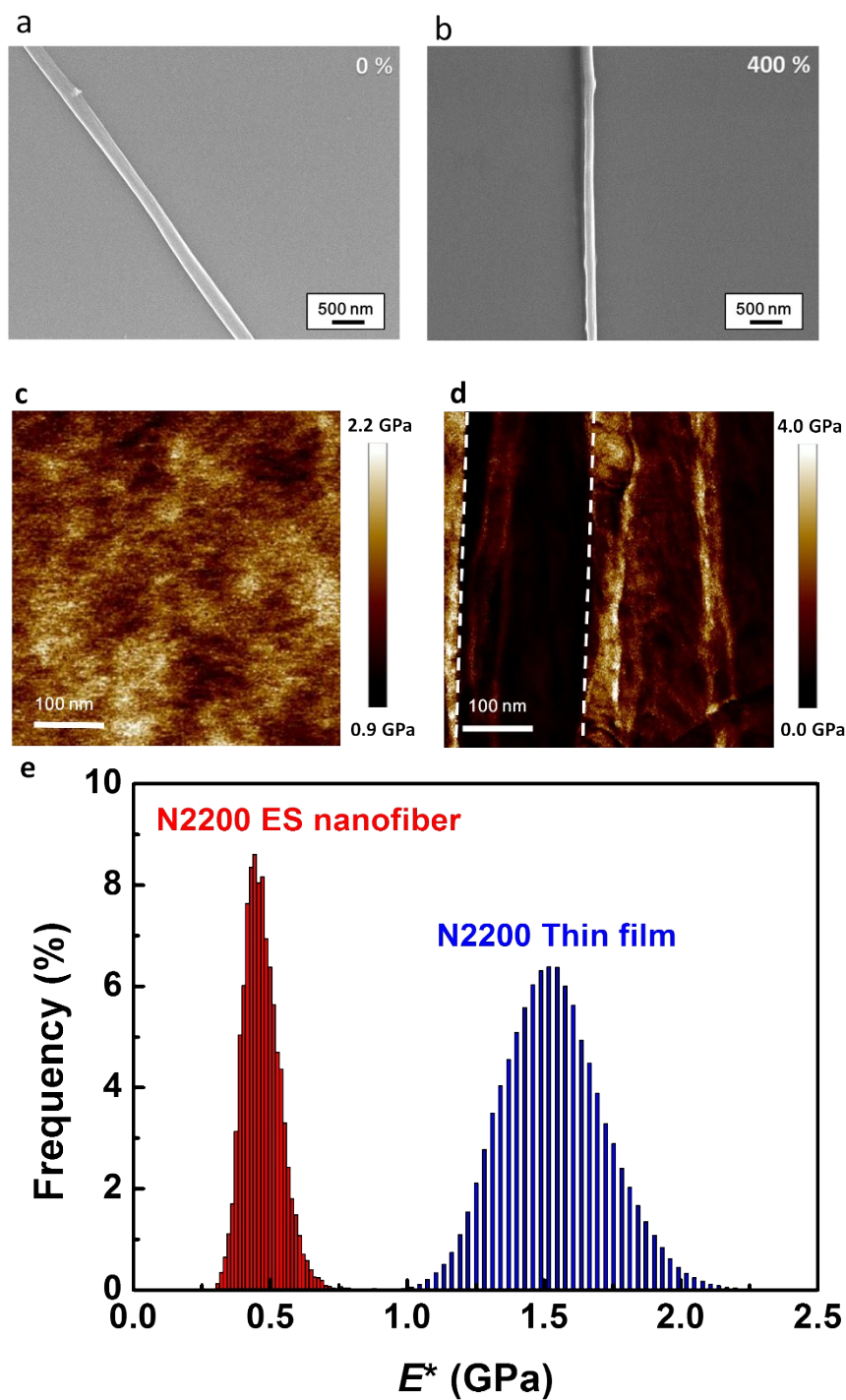
**Fig. S2** SEM images of (a) core-shell P3HT/PEO ES fibers (a) before solvent etching and (b) after solvent etching. Confocal images of core-shell P3HT/PEO ES fibers (c) before solvent etching and (d) after solvent etching. Noted that the flattened core-shell P3HT/PEO ES fiber may result from the mechanical stress from cover slip.



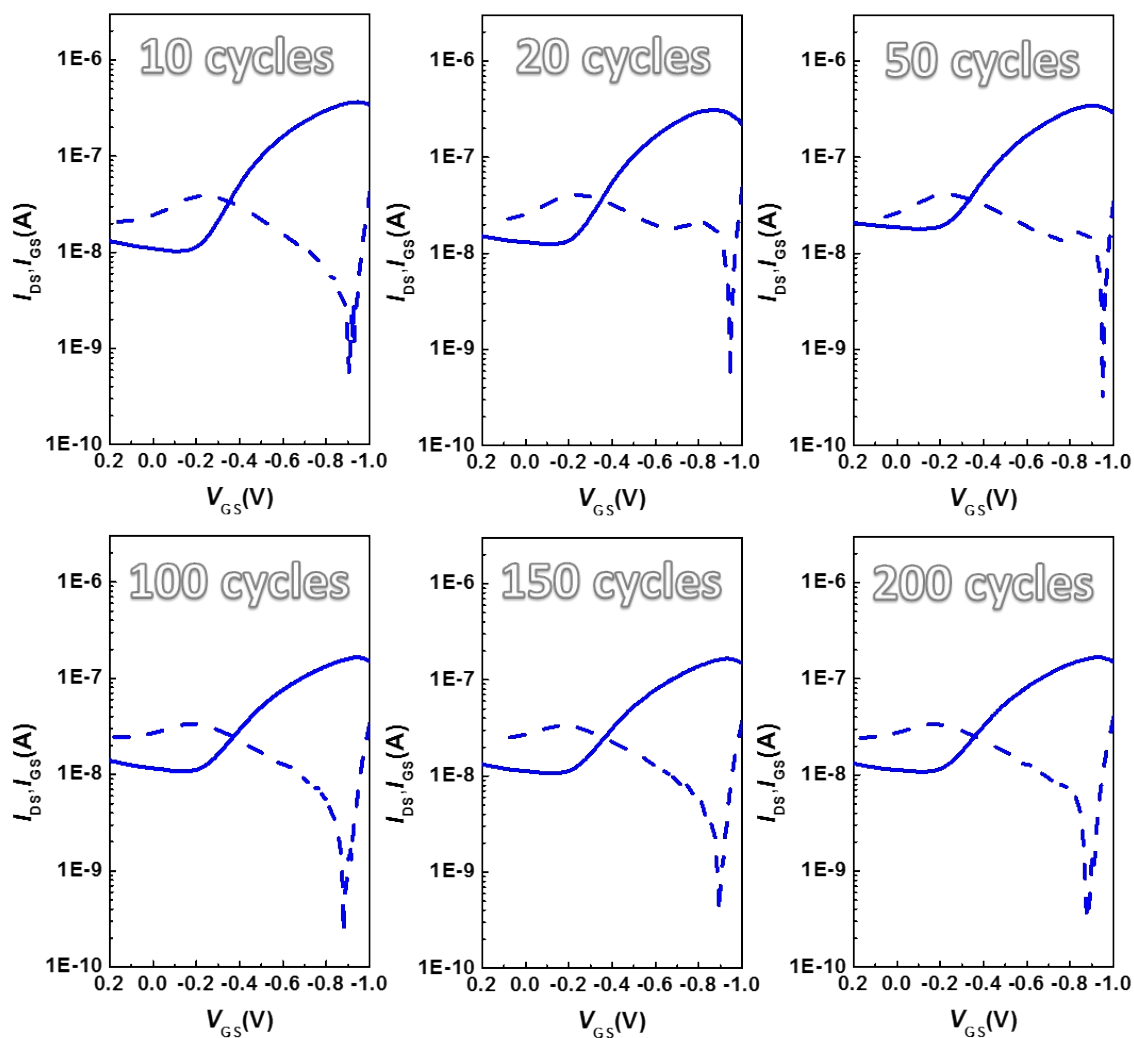
**Fig. S3** (a) SEM image of P3HT ES nanofiber and the corresponding EDS mapping of (b) sulfur atom and (c) oxygen atom. (d) The analysis of element composition on the selected area of P3HT ES nanofiber shows 5.34 % of sulfur, 83.94 % of carbon and 10.72 % of silicon. Noted that very light red color representing oxygen atom in (c) is the cause of native oxide layer of ca. 2 nm on bare silicon wafer.



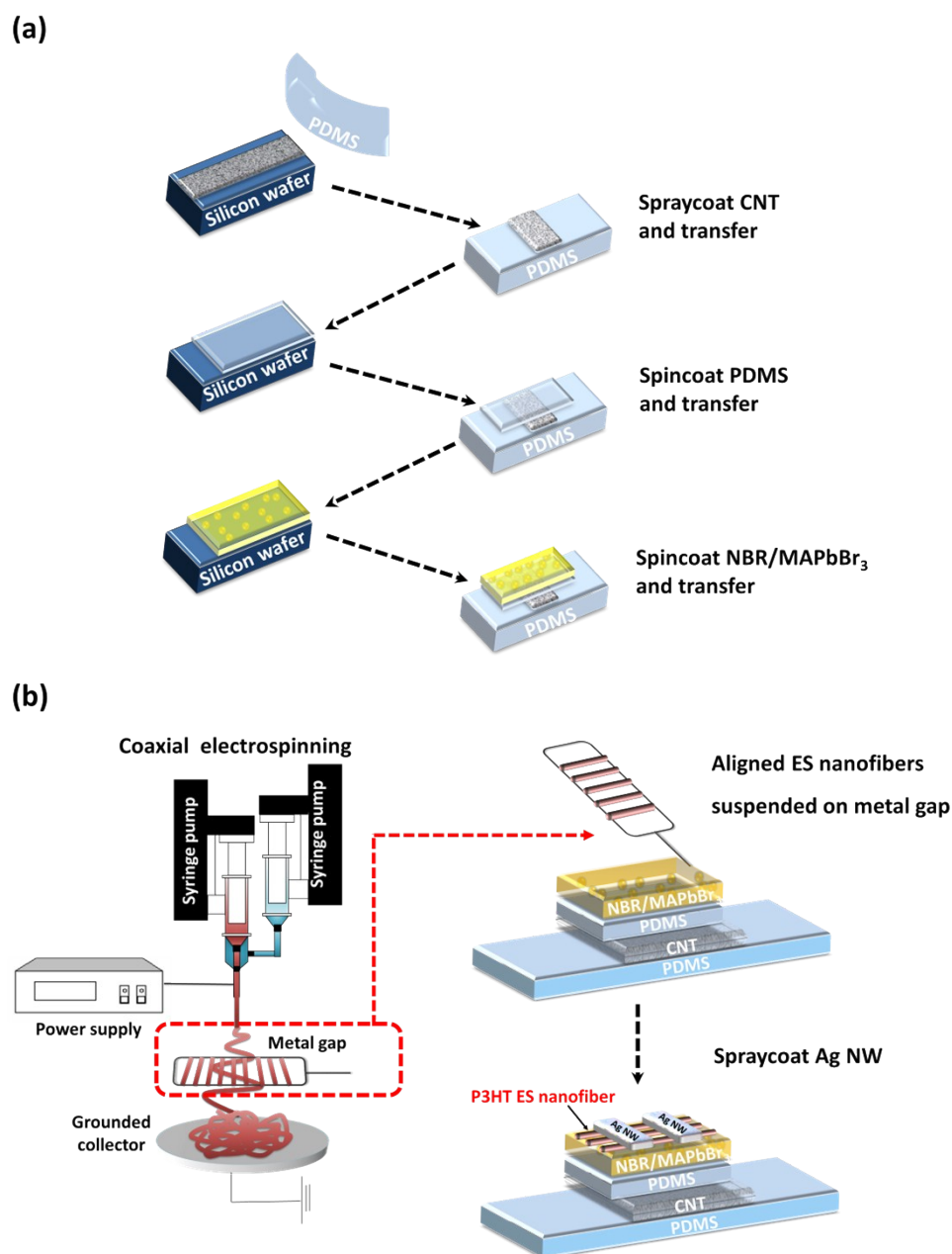
**Fig. S4** Optical microscope images of the studied polymer films under the strain of (a) 0 %, (b) 25 %, (c) 50 % and (d) 75 % for P3HT film; (e) 0 %, (f) 25 %, (g) 50 %, (h) 75 % for N2200 film. The scale bar is 10  $\mu\text{m}$ . DSC thermograms of (i) pristine P3HT and P3HT ES nanofibers and (j) second scan of P3HT ES nanofibers.



**Fig. S5** SEM images of N2200 ES nanofiber under strain of (a) 0 % and (b) 400 %. DMT modulus mapping of (c) N2200 ES nanofiber and (d) N2200 thin film. (e) Reduced modulus distributions of N2200 ES nanofiber and N2200 thin film, respectively.

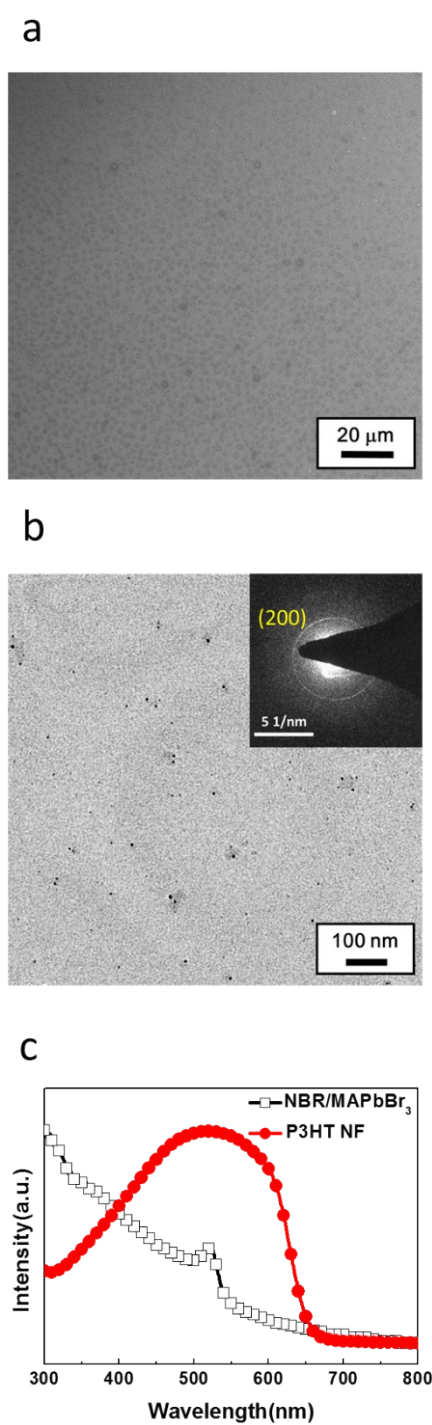


**Fig. S6** Transfer curves of fully stretchable P3HT ES nanofibers-based FET at 0% strain after multiple stretching-releasing cycles between 0 % and 100 % strain.

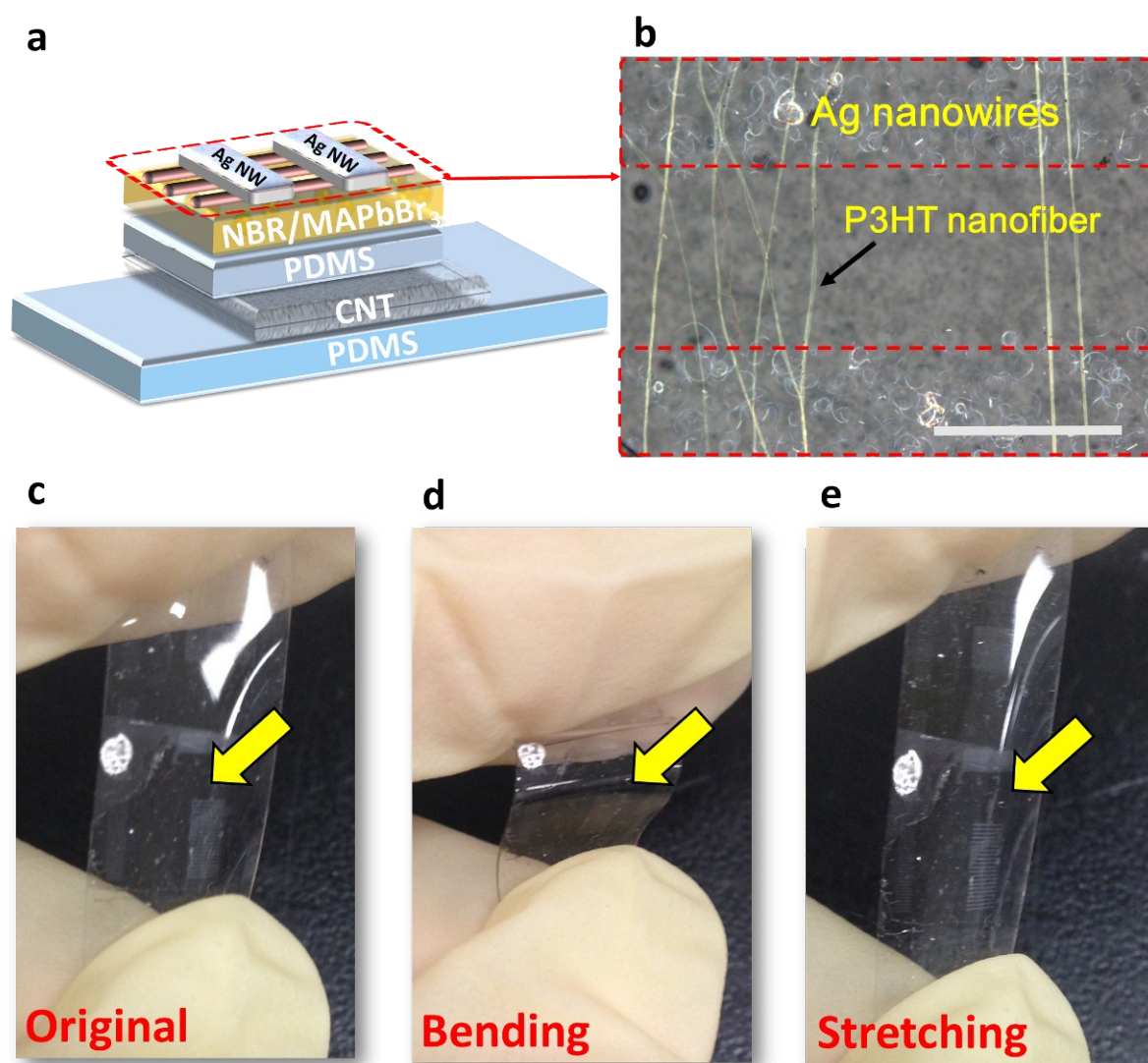


**Fig. S7** Schematic illustration of preparation process for stretchable perovskite-based photomemory: (a) fabrication of stretchable PDMS substrate, gate electrode, gate dielectric layer and photoactive layer; (b) preparation of P3HT ES nanofibers and assemble the fully stretchable perovskite-based photomemory.





**Fig. S8** (a) SEM image of MAPbBr<sub>3</sub>/NBR composite film. (b) TEM image of MAPbBr<sub>3</sub>/NBR composite film with the inserted SAED image. (c) Optical absorption of P3HT nanofibers and MAPbBr<sub>3</sub>/NBR composite film.



**Fig. S9** (a) Schematic of fully stretchable photomemory. (b) Bright-field optical images of the device (scale bars = 100  $\mu\text{m}$ ). (c-e) The images of fully stretchable device on original state, under bending and stretching where the arrows indicate the position of devices.

**Table S1.** Summarized electrical performance of stretchable FETs under strain reported in the literature so far.

<b>Stretchable Semiconducting Material</b>	<b>Approach</b>	<b>Mobility at <math>\epsilon</math> of 0% (<math>\text{cm}^2/\text{Vs}</math>)</b>	<b>Mobility under strain (<math>\text{cm}^2/\text{Vs}</math>)</b>	<b>Modulus variation</b>
<b>DPP-polymer (ref: Nature 2016, 539, 411)</b>	Dynamic Bonding (Chemistry)	1.32	$1.10 \times 10^{-1}$ at $\epsilon$ of 100 %	2.19 GPa (DPP-polymer) $\rightarrow$ 341 MPa (DPP-polymer with 10% PDCA)
<b>DPP polymer/SEBS (ref: Science 2017, 549, 59)</b>	Composite (Physics)	$\sim 1.08$	1.08 at $\epsilon$ of 100 %	$\sim 750$ MPa (DPPT-polymer thin film) $\rightarrow \sim 50$ MPa (DPP-polymer/SEBS composite film)
<b>P3HT nanofibril/SEBS (ref: Adv. Mater. 2015, 27, 1255)</b>	Composite (Physics)	$\sim 6.00 \times 10^{-3}$	$\sim 2.00 \times 10^{-3}$ at $\epsilon$ of 50 %	11.7 MPa (P3HT/ SEBS composite film)
P3HT(This Work)	<b>Electrospinning (Physics)</b>	<b>2.4</b>	<b><math>1.59 \times 10^{-1}</math> at <math>\epsilon</math> of 120 %</b>	<b>3.93 GPa (P3HT thin film) <math>\rightarrow</math> 448 MPa (P3HT ES nanofiber)</b>

**Table S2.** Electrical properties of fully stretchable P3HT ES nanofibers-based FET under strain within 0-120 %.

<b>Strain (%)</b>	<b>Mobility (cm<sup>2</sup> V<sup>-1</sup>s<sup>-1</sup>)</b>	<b>ON/OFF current ratio (-)</b>	<b>V<sub>th</sub> (V)</b>
0	2.40	1.67×10 <sup>3</sup>	-0.10
20	1.87	2.71×10 <sup>2</sup>	-0.05
40	1.38	3.29×10 <sup>2</sup>	-0.01
60	9.56×10 <sup>-1</sup>	1.52×10 <sup>3</sup>	-0.06
80	9.35×10 <sup>-1</sup>	2.75×10 <sup>3</sup>	-0.08
100	7.15×10 <sup>-1</sup>	1.13×10 <sup>3</sup>	-0.10
120	1.59×10 <sup>-1</sup>	1.35×10 <sup>3</sup>	-0.13
release	4.30×10 <sup>-1</sup>	5.22×10 <sup>3</sup>	-0.14

**Table S3.** Electrical properties of fully stretchable P3HT ES nanofibers-based FET at 0% strain after multiple stretching(100 % strain)-releasing cycles.

<b>Cycle</b>	<b>Mobility (cm<sup>2</sup> V<sup>-1</sup>s<sup>-1</sup>)</b>	<b>ON/OFF current ratio (-)</b>	<b>V<sub>th</sub> (V)</b>
1	4.30×10 <sup>-1</sup>	5.22×10 <sup>3</sup>	-0.14
10	2.00×10 <sup>-1</sup>	3.58×10 <sup>1</sup>	-0.10
20	1.51×10 <sup>-1</sup>	2.49×10 <sup>1</sup>	-0.11
50	2.08×10 <sup>-1</sup>	1.92×10 <sup>1</sup>	-0.09
100	1.31×10 <sup>-1</sup>	1.30×10 <sup>1</sup>	-0.06
150	7.23×10 <sup>-2</sup>	1.53×10 <sup>1</sup>	-0.10
200	8.58×10 <sup>-2</sup>	1.55×10 <sup>1</sup>	-0.04