Supporting Information

Electrically Activated Paper Actuators

Mahiar M. Hamedi [†] Victoria E. Campbell,[†] Philipp Rothemund, Firat Güder, Dionysios C. Christodouleas, Jean-Francis Bloch, George M. Whitesides*

Dr. M. M. Hamedi, Dr. V. E. Campbell, P. Rothemund, Dr. F. Guder, Dr. D. C. Christodouleas, Prof. G. M. Whitesides
Department of Chemistry and Chemical Biology, Harvard University, Cambridge MA, USA
[†]Both authors contributed equally to this work.
E-mail: gwhitesides@gmwgroup.harvard.edu

Dr. V. E. Campbell
Institut de Chimie Moléculaire et des Matériaux d'Orsay, CNRS, Université Paris Sud 11, 91405 Orsay Cedex, France.

P. Rothemund
School of Engineering and Applied Sciences, Harvard University, Cambridge, MA 02138, USA.

P. Rothemund, Prof. G. M. Whitesides
Kavli Institute for Bionano Science and Technology, Harvard University, Cambridge MA 02138, USA.

Prof. J.-F. Bloch
Grenoble Institute of Technology, The International School of Paper, Print Media and Biomaterials (PAGORA), France

Prof. G. M. Whitesides
Wyss Institute for Biologically Inspired Engineering, Harvard University, Cambridge, MA, USA

An analytical solution for the bending curvature of a bi-layer actuator

We assume that the layers in the paper/PEDOT:PSS composite have a linear elastic behavior, so that their material law can be written as

$$\varepsilon = \frac{\sigma}{E_i} + \varepsilon_{hi}, \tag{1}$$

Where, σ is the axial stress, E_i the Young's modulus and ε_{hi} the strain caused by electrothermal heating. The strain energy per volume of the material can therefore be derived as

$$U_i = \int_0^{\varepsilon} \sigma \, d\varepsilon = \frac{1}{2} E_i \varepsilon^2 + E_i \varepsilon \varepsilon_{hi} \tag{2}$$

We model the HEPAs as an Euler-Bernoulli beam. We assume perfect bonding between paper (thickness t_1, Young's modulus E_1, hygroexpansive strain $\varepsilon_{h1} = \varepsilon_h$) and the strain limiting layer (thickness t_2, Young's modulus E_2, no hygroexpansive strain ($\varepsilon_{h2} = 0$)), so that the strain across the thickness of the actuator is linear:

$$\varepsilon = \varepsilon_0 - \kappa y \tag{3}$$

$\kappa = 1/r$ is the curvature of the actuator (Figure S3a), y is a coordinate in direction of the thickness of the HEPA with arbitrary origin.

$$\Phi = \iiint_{Volume} \frac{1}{2} E(y) \varepsilon(y)^2 + E(y) \varepsilon(y) \varepsilon_h(y) \, dV \tag{4}$$

To obtain a formula for the curvature of the bending actuator the energy of deformation of the whole actuator has to be minimized with respect to κ and ε_0. In equation (4) $E(y)$ is E_1 in the paper/PEDOT:PSS composite layer and E_2 in the strain limiting layer. $\varepsilon_h(y)$ is ε_h in the paper layer and 0 in the strain limiting layer. Using $\varepsilon(y)$ from equation 3 leads to

$$\kappa = \frac{1}{r} = \frac{6 \varepsilon_h E_1 E_2 t_1 t_2 (t_1 + t_2)}{(E_1 t_1^2 - E_2 t_2^2)^2 + 4 E_1 E_2 t_1 t_2 (t_1 + t_2)^2} \tag{5}$$

The result is analogous to a bi-layered beam under thermal expansion.[1]

Experimental calculation of the change in strain

We calculated that ε_h = 0.3% using equation (5), by taking the largest measured curvature of κ = 0.12 cm^{-1} for the actuator from Figure S2. In this calculation we used the following values for Young's modulus and thickness: E_1 = 1.6 GPa (for the paper, based on the average value from experimental results measured at RH=40%), E_2 = 2.0 GPa (table values for polyethylene terephthalate), t_1 = 180 µm, t_2 = 51 µm.

We considered the detailed relation between the change in mechanical and structural properties of the single fibers, and that of the entire paper/PEDOT:PSS composite structure, which depends on the structure of the network, and the interactions between mechanical properties and hygroexpansion.[2,3]

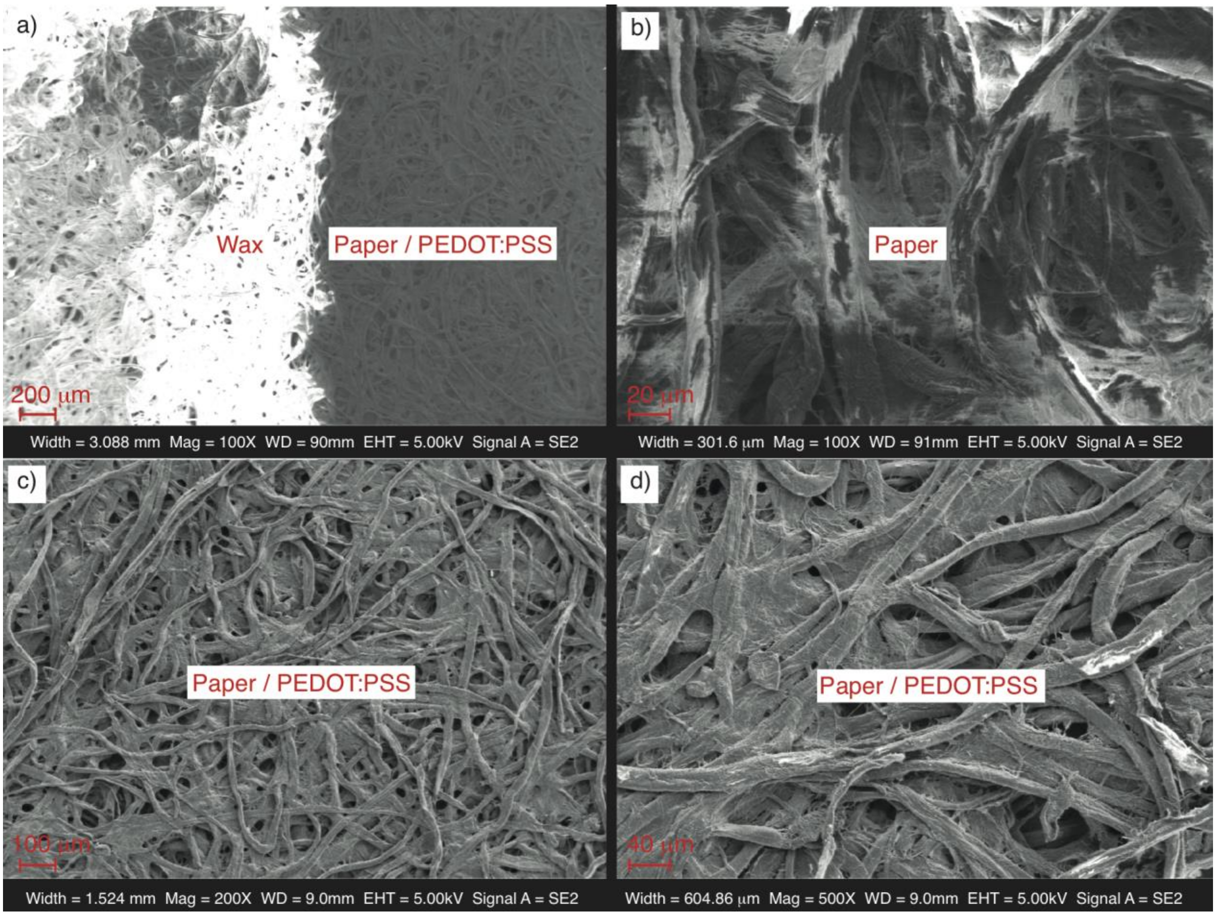

Figure S1. Scanning electron microscopic (SEM) images of (a) the wax / paper /PEDOT:PSS composite interface, (b) the paper, and (c and d) the paper/PEDOT:PSS composite at different magnifications.

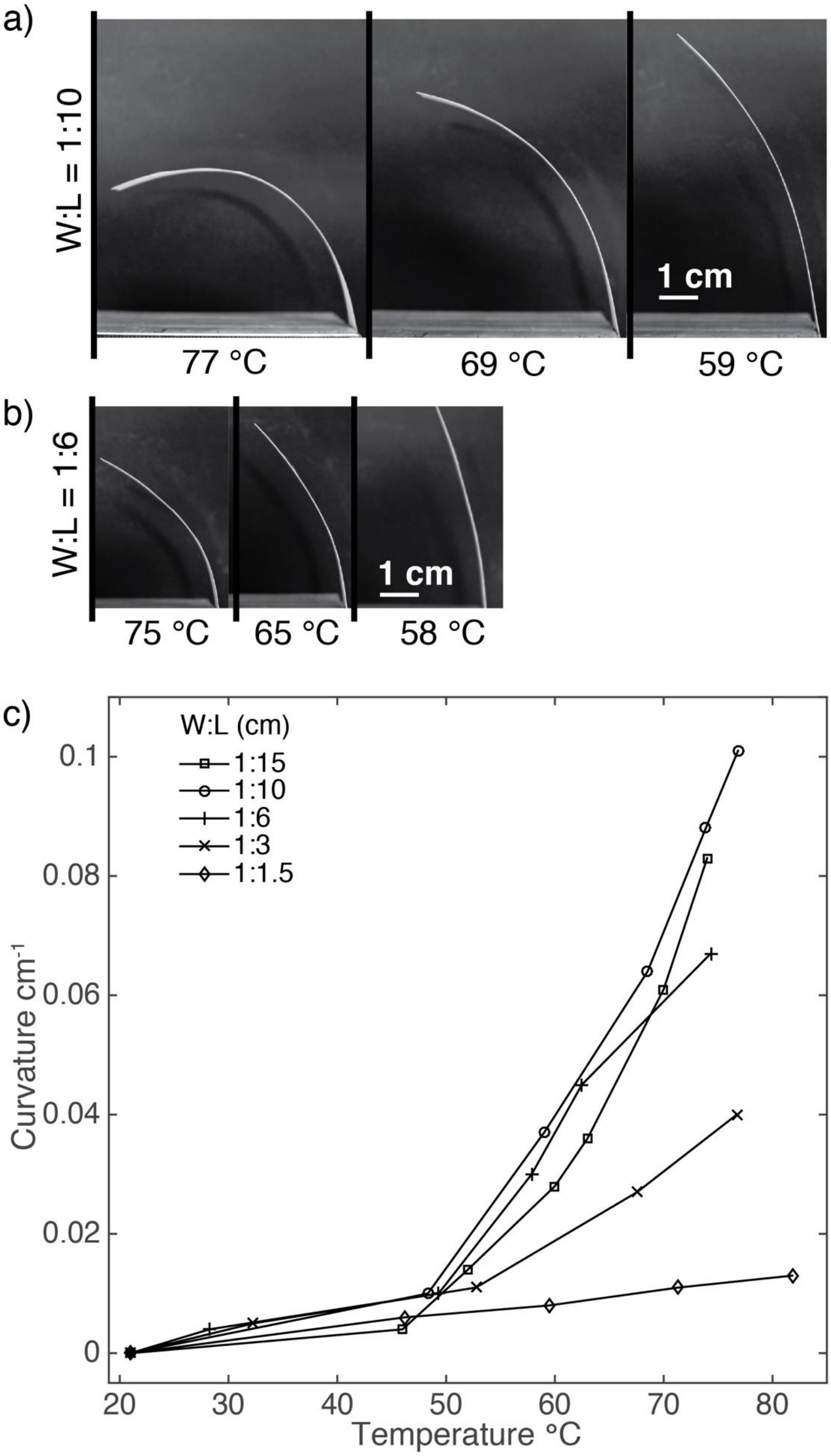

Figure S2. Photographic images of the actuators at different operating temperature/power, with two different L:W ratios: a) L:W = 10:1, and b) L:W = 6:1 (RH = 40%; c) Curvature vs.

temperature measured for actuators with different length/width ratio. The temperature was varied by varying the driving power of the actuators, and recorded at equilibrium with an IR camera. The curvature was calculated from photos of the actuators. The actuators moved parallel to the gravitational force, so that we could neglect gravity.

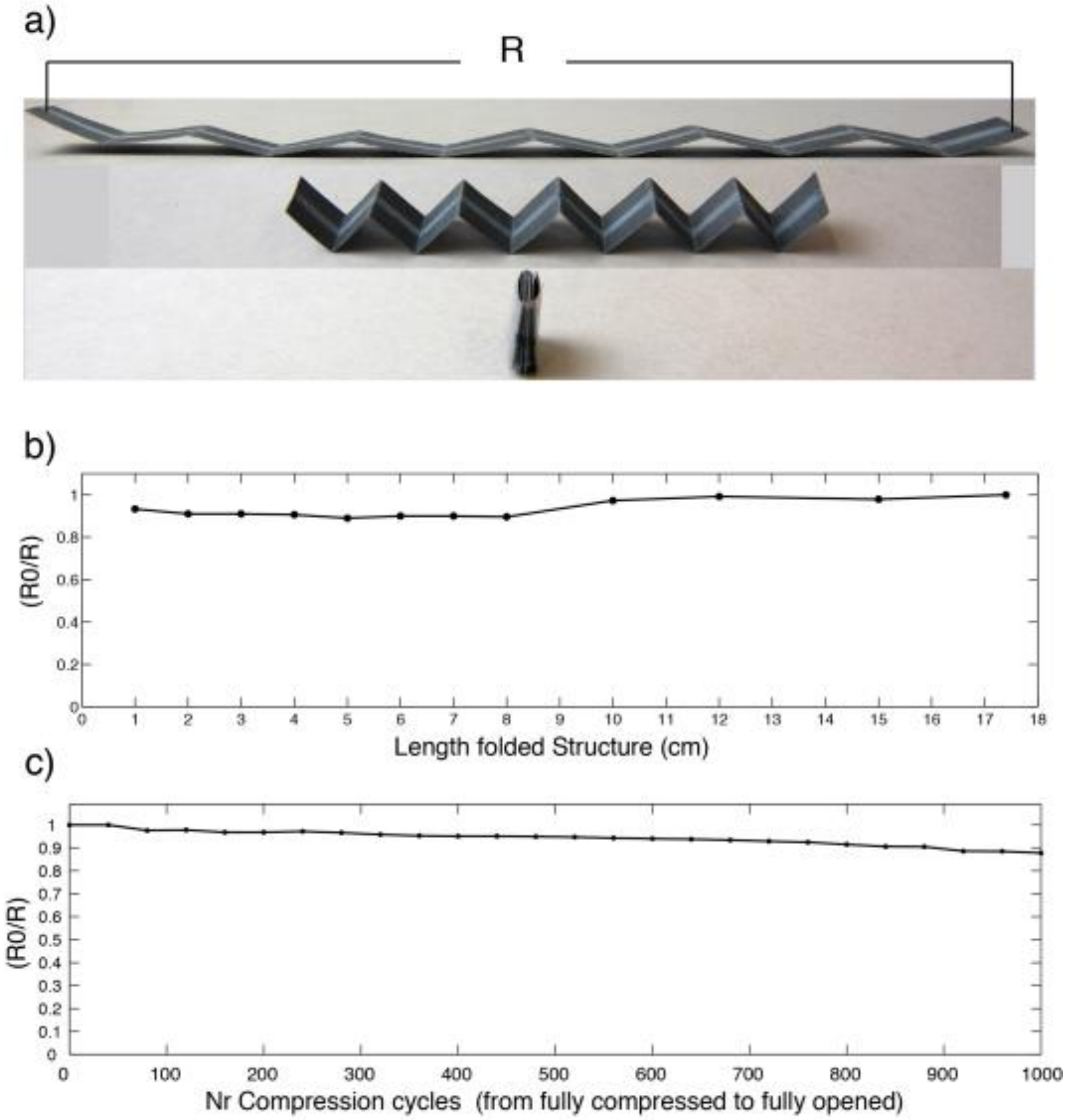

Figure S3. a) Photographs of an accordion shaped folded paper structure at different compression lengths. b) Normalized resistance of the paper electrode measured from one side of the folded paper to the other side, as a function of compressed length. c) Normalized resistance (R/R_0) measured at the uncompressed state, as a function of number of cycles, where each cycle corresponds to a complete closing and opening of the folded structure (i.e. moving each crease from 180 to 0 degrees angle).

Supporting Information References

[1] S. Timoshenko, *J. Opt. Soc. Am.* **1925**, *11*, 233.

[2] W. W. Sampson, J. Yamamoto, *J. Mater. Sci.* **2010**, *46*, 541.

[3] T. Uesaka, *J. Mater. Sci.* **1994**, *29*, 2373.