Supporting Information

Electrically Activated Paper Actuators

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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

\[
e = \frac{\sigma}{E_i} + \varepsilon_{hi} \tag{1}\]

Where, \(\sigma\) is the axial stress, \(E_i\) the Young’s modulus and \(\varepsilon_{hi}\) the strain caused by electrothermal heating. The strain energy per volume of the material can therefore be derived as

\[
U_i = \int_0^d \frac{1}{2} E_i \varepsilon^2 + E_i \varepsilon \varepsilon_{hi} \, \mathrm{d}V \tag{2}
\]

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:

\[
\varepsilon = \varepsilon_0 - k y \tag{3}
\]

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

\[
= \int_{\text{Volume}} \frac{1}{2} E(y) (y')^2 + E(y) \varepsilon(y) \varepsilon_{h}(y) \, \mathrm{d}V \tag{4}
\]

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 \(\kappa\) and \(\varepsilon_0\). 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 \(\varepsilon_h\) in the paper layer and 0 in the strain limiting layer. Using \(\varepsilon(y)\) from equation 3 leads to

\[
\kappa = \frac{6 E_1 E_2 t_1 t_2 (t_1 + t_2)}{r \left( E_1 t_1^2 + E_2 t_2^2 \right)^2 + 4 E_1 E_2 t_1 t_2 (t_1 + t_2)^2} \tag{5}
\]

The result is analogous to a bi-layered beam under thermal expansion.[1]
Experimental calculation of the change in strain

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

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.\textsuperscript{[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

