Supporting Information

Self-Standing Cellulose Nanofibers/ Poly(3,4-ethylenedioxythiophene):

Poly (4-styrenesulfonate)/Ionic Liquid Actuators with Superior Performance

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Table S1: Specific capacitance, $C = C_1$, divided by the weights of PEDOT:PSS (F g^{-1}) in CNF/PEDOT:PSS/IL electrodes (applied triangular voltage: ±0.5 V, sweep rate: 1 mV s^{-1}).

IL	LB/ PEDOT	BB/ PEDOT	LB/ PEDOT	BB/ PEDOT
	50 200 200	50 200 200	50 100 200	50 100 200
EMI[BF$_4$]	24.0	24.1	27.4	23.8
EMI[CF$_3$SO$_3$]	26.5	26.3	26.4	24.2

Table S2: Electrical conductivity (S cm^{-1}) of CNF/PEDOT:PSS/IL electrodes.

IL	LB/ PEDOT	BB/ PEDOT	LB/ PEDOT	BB/ PEDOT
	50 200 200	50 200 200	50 100 200	50 100 200
EMI[BF$_4$]	6.3	5.0	5.6	4.5
EMI[CF$_3$SO$_3$]	4.8	4.8	5.9	6.6

Table S3: Maximum strain (%) of CNF/PEDOT:PSS/IL (50/100/200) electrodes.

IL	LB/ PEDOT	BB/ PEDOT
EMI[BF_4]	0.45	0.72
EMI[CF_3SO_3]	0.39	0.46

Table S4: Maximum strain (%) of CNF/PEDOT:PSS/IL (50/200/200) and PEDOT:PSS/IL electrodes.

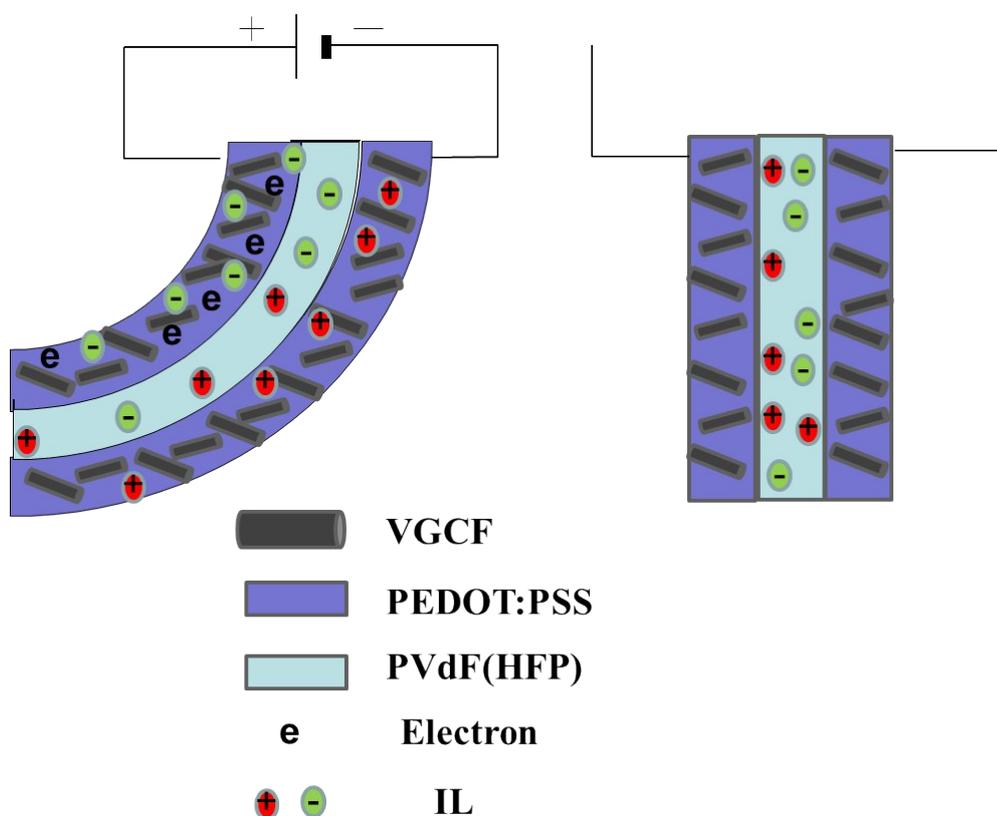| IL	LB/PEDOT	BB/ PEDOT	PEDOT
EMI[BF_4]	0.74	1.04	0.26*
EMI[CF_3SO_3]	0.64	0.78	0.48*

* Ref. [33]

Table S5 Maximum generated stress values (MPa) of the (50/200/200) and PEDOT:PSS/IL electrodes.
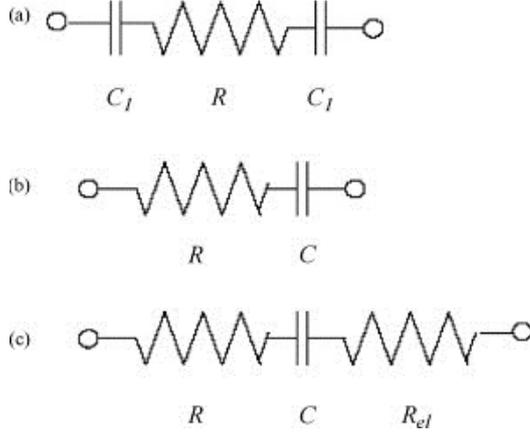
IL	LB/PEDOT	BB/ PEDOT	PEDOT
EMI[BF$_4$]	1.02	1.41	0.35*
EMI[CF$_3$SO$_3$]	0.90	1.22	0.49*

* Ref. [33]

VGCF

PEDOT:PSS

PVdF(HFP)

e   **Electron**

IL

Figure S1: Schematic of the response model used for the PEDOT:PSS/VGCF/IL actuators.

Figure S2: Three equivalent circuit models for the CNF actuator, where C_1 and C are specific resistances (with $C = C_1/2$), R is the ionic resistance, *and R_{el} is the electrode resistance.*

Figure S2 presents equivalent circuit models for CNF/PEDOT:PSS/IL, actuators. The model in **Figure S2(a)** consists of the specific capacitance C_1 between the CNF/PEDOT:PSS/IL electrode and the electrolyte layer; and the resistance, *R, associated with* the electrolyte layer. **Figure S2(b)** shows a more simplified model in which the two C_1 capacitances are replaced by a single capacitance C (= $C_1/2$). When a triangular voltage with an amplitude of $\pm A$ and frequency of f is applied to the equivalent circuit shown in **Figure S2(b)**, the maximum accumulated charge $Q(f)$ can be expressed as follows [S1]:

$$Q(f)/Q_0 = 1 - 4CRf\,(1 - \exp(-1/4CRf)), \qquad (S1)$$

where Q_0 is the accumulated charge at the low-frequency limit. If the strain ε in the electrode layer is proportional to the accumulated charge, then it can be calculated as follows:

$$\varepsilon = \varepsilon_0\, Q(f)\,/\,Q_0, \qquad (S2)$$

where ε_0 is the strain at the low-frequency limit.

When conduction is considered in the electrode layer, it is necessary to account for the electrode resistance in the equivalent circuit. If the electrode resistance is treated explicitly, then the equivalent circuit should be treated as a distributed transmission line [S2]. Here, we assumed that the electrode resistance consists of a resistance element, R_{el}, as shown in **Figure S2(c)**. Thus, R in Eq. (S1) can be replaced by $R + R_{el}$.

To evaluate the double-layer charging kinetic model, which accounts for the oxidization and reduction reactions of the PEDOT, the specific capacitances of the CNF/PEDOT:PSS/IL electrodes were measured along with the ionic resistance of the gel electrolyte layer. The frequency dependence of the strain was calculated using Equation (S1) and Equation (S2). **Figure S3** shows the frequency dependence of the measured strain values together with the simulation results for the BB/PEDOT:PSS/EMI[BF$_4$] (50/200/200), actuators. Curve A was calculated using the model shown in **Figure S2(b)**, whereas **Table S6** lists the simulation parameters. Curve B was calculated using the model shown in **Figure S2(c)**, and the corresponding simulation parameters are listed in **Table S7**. **Figure S3** clearly shows that the frequency dependence of the strain is closely reproduced by Curve B. **Figure S3** further reveals that the frequency dependence of the strain is reproduced by the double-layer charging kinetic model when the electrode resistance is considered. Similar results were obtained for the LB/PEDOT:PSS/EMI[BF$_4$] (50/200/200), CNF/PEDOT:PSS/EMI[CF$_3$SO$_3$] (50/200/200) and CNF/PEDOT:PSS/IL (50/100/200) actuators.

The strain values were fitted to the low-frequency limit, as shown in Figure **S3**, by choosing appropriate values for ε_0 in Eq. (S2). These are listed in **Table S7**.

The results summarized in **Table S6** and **Table S7** should be considered with respect to both the kinetic and static components to obtain an actuator with optimal performance. From a kinetic viewpoint, the most important consideration is that the frequency dependence of the strain is determined by electrochemical charging, as shown in **Figure S3.** Thus, obtaining good fits generally requires taking the electrode resistance into account, and the responses of the CNF/PEDOT:PSS/IL actuators can be improved by fabricating electrodes with higher conductivities.
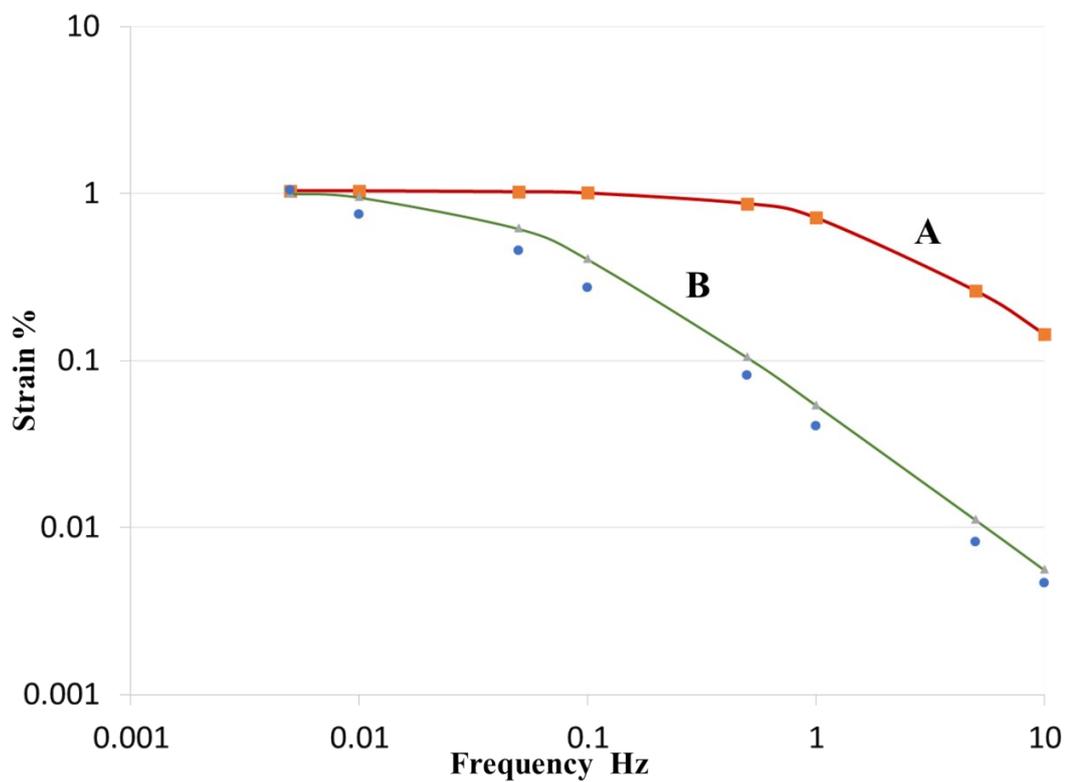
Figure S3: Measured (blue dots and orange rectangular symbols) and simulated (green and red curves) data showing the frequency dependence of the strain for a BB/PEDOT:PSS/EMI[BF$_4$] (50/200/200) device. Curves A and B were calculated using the equivalent circuits in **Figures S2(b)** and **S2(c)**, respectively.

Table S6: Simulation parameters for the CNF/PEDOT:PSS/IL (50/200/200) model that ignores electrode resistance.

CNF/IL	C_{PEDOT} (F g^{-1})	C (F cm^{-2})	κ (mS cm^{-1}) [a]	R (Ω cm^2)	ε_0 (%)	CR (s)
LB/[BF$_4$]	24.0	0.2803	2.8	0.714	0.74	0.2001
BB/[BF$_4$]	24.1	0.1807	2.8	0.714	1.04	0.1290
LB/[CF$_3$SO$_3$]	26.5	0.2657	4.4	0.455	0.64	0.1209
BB/[CF$_3$SO$_3$]	26.3	0.2488	4.4	0.455	0.78	0.1132

[a] ref. [S3]

Table S7: Simulation parameters for the CNF/PEDOT:PSS/IL (50/200/200) model that considers electrode resistance.

IL	C (F cm^{-2})	R_{el} (Ω cm^2)	$R+R_{el}$ (Ω cm^2)	$C(R+R_{el})$ (s)
LB/[BF$_4$]	0.2803	10.6	11.3	3.17
BB/[BF$_4$]	0.1807	12.5	13.2	2.39
LB/[CF$_3$SO$_3$]	0.2657	13.4	13.9	3.69
BB/[CF$_3$SO$_3$]	0.2488	13.9	14.4	3.58

R_{el} = area of the electrode film (cm^2)/[electrical conductivity (S cm^{-1}) $\times$ thickness of the electrode film (cm)] [S1].

References

[S1] I. Takeuchi, K. Asaka, K. Kiyohara, T. Sugino, K. Mukai, T. Fukushima and T. Aida, *Electrochim. Acta*, 2009, **53**, 1762-1768.

[S2] K. Takagi, Y Nakabo, Z.-W. Luo and K. Asaka, *Proceedings of the SPIE, Electroactive Polymer Actuator and Devices (EAPAD)*, 2007, **6524**, 652416-1-652416-8.

[S3] N. Terasawa, I. Takeuchi and H. Matsumoto, *Sens. Actuators B*, 2009, **139,** 624-630.