Supporting Information

Performance Enhancement of PEDOT:Poly(4-styrenesulfonate) Actuators by Using Ethylene Glycol

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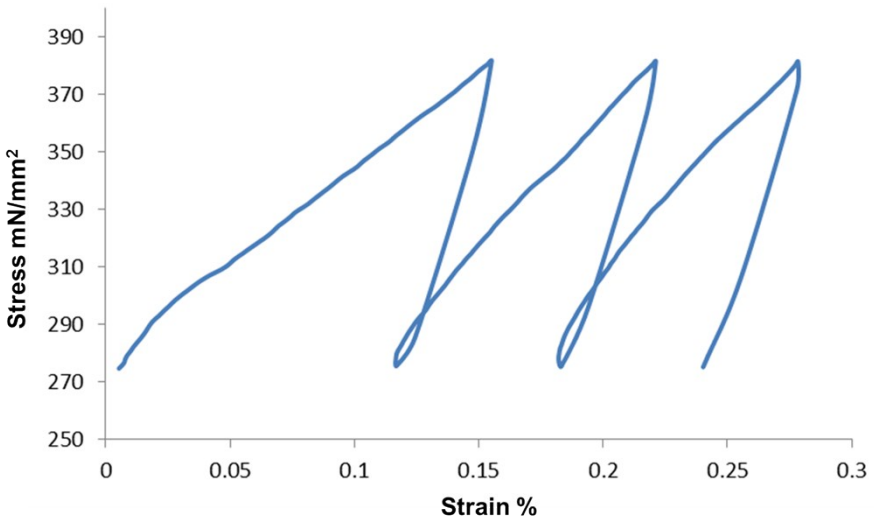
a)



b)

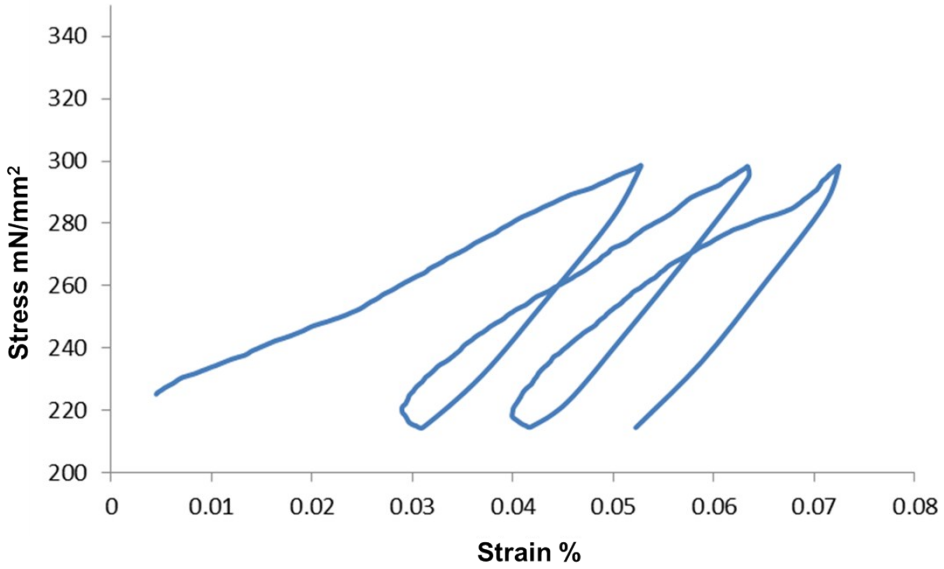


Figure S1 Stress (**mN mm^{-2}**) vs. strain (**%**) curve for a) PEDOT:PSS/VGCF/EMI[BF_4]/EG 30 % and b) PEDOT:PSS/VGCF/EMI[BF_4] electrodes.

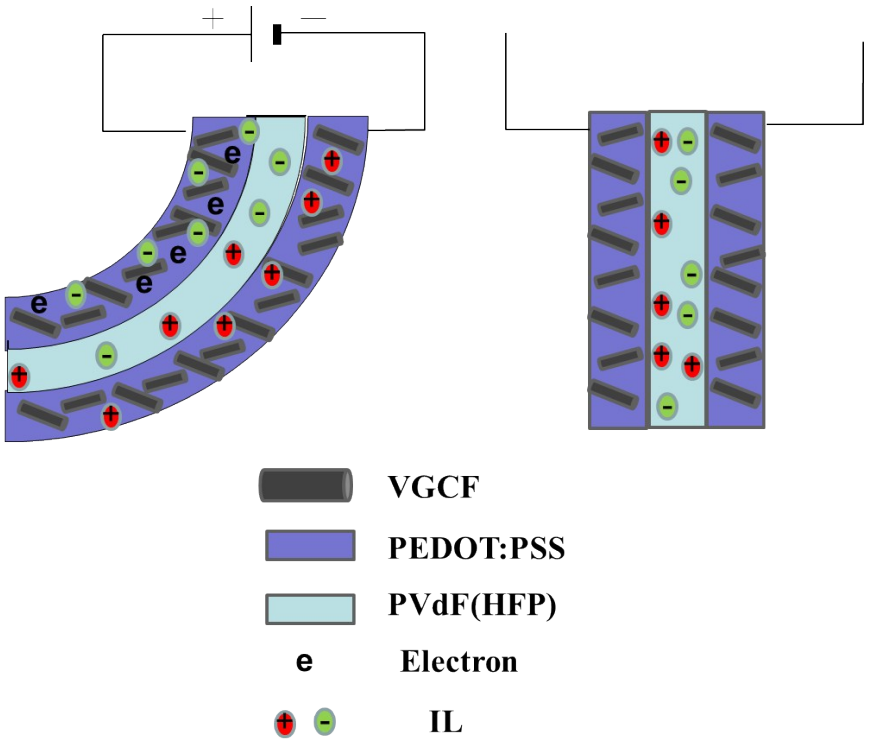


Figure S2 Schematic of the response model used for PEDOT:PSS/VGCF/IL actuators.

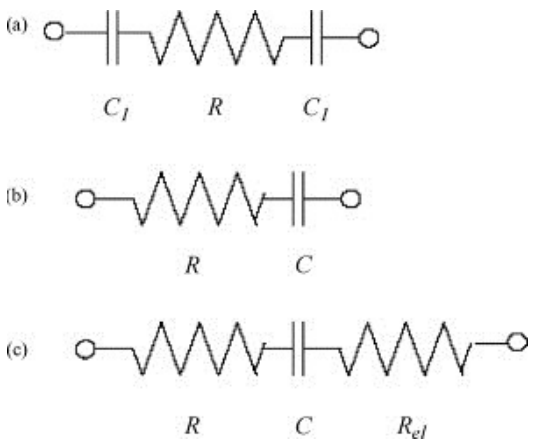


Figure S3 Three equivalent circuit models for the bucky-gel actuator. C_1 and C: specific resistances (with $C = C_1/2$), R: ionic resistance, R, and R_{el}: electrode resistance.

Figure S3 presents equivalent circuit models for PEDOT:PSS/VGCF/IL, actuators. The model in Figure S3(a) consists of the specific capacitance C_1 between the PEDOT:PSS/VGCF/IL electrode and the electrolyte layer; and the resistance, R, associated with the electrolyte layer. Figure S3(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 S3(b), the maximum accumulated charge $Q(f)$ can be expressed as follows [S1]:

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

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:

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

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

When conduction is considered in the electrode layer, the electrode resistance must be accounted for 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 S3(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 PEDOT:PSS/VGCF/IL/EG 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 S4** show the frequency dependence of the measured strain values together with simulation results for the PEDOT:PSS/VGCF/EMI[CF_3SO_3]/EG 3%, actuators. Curve A was calculated using the model shown in Figure S3(b), while **Table S1** list the simulation parameters. Curve B was calculated using the model shown in Figure S3(c), and the corresponding simulation parameters are listed in **Table S2**. Figure S4 clearly show that the frequency dependence of the strain is well-reproduced by Curve B. Figure S4 further reveal 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 PEDOT:PSS/VGCF/EMI[CF_3SO_3]/EG 30%, PEDOT:PSS/VGCF/EMI[BF_4]/EG and PEDOT:PSS/VGCF/IL actuators.

To fit the strain values in the low-frequency limit, as shown in Figure S4, appropriate values were chosen for ε_0 in Eq. (S2). These ar0e listed in Table S2.

To optimize the performance of the actuator, the results summarized in Table S1 and Table S2 should be considered with respect to both the kinetic and static components. 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 S4. Thus, obtaining good fits generally requires taking the electrode resistance into account, and the responses of the PEDOT:PSS/VGCF/IL/EG actuators can be improved by fabricating electrodes with higher conductivities.

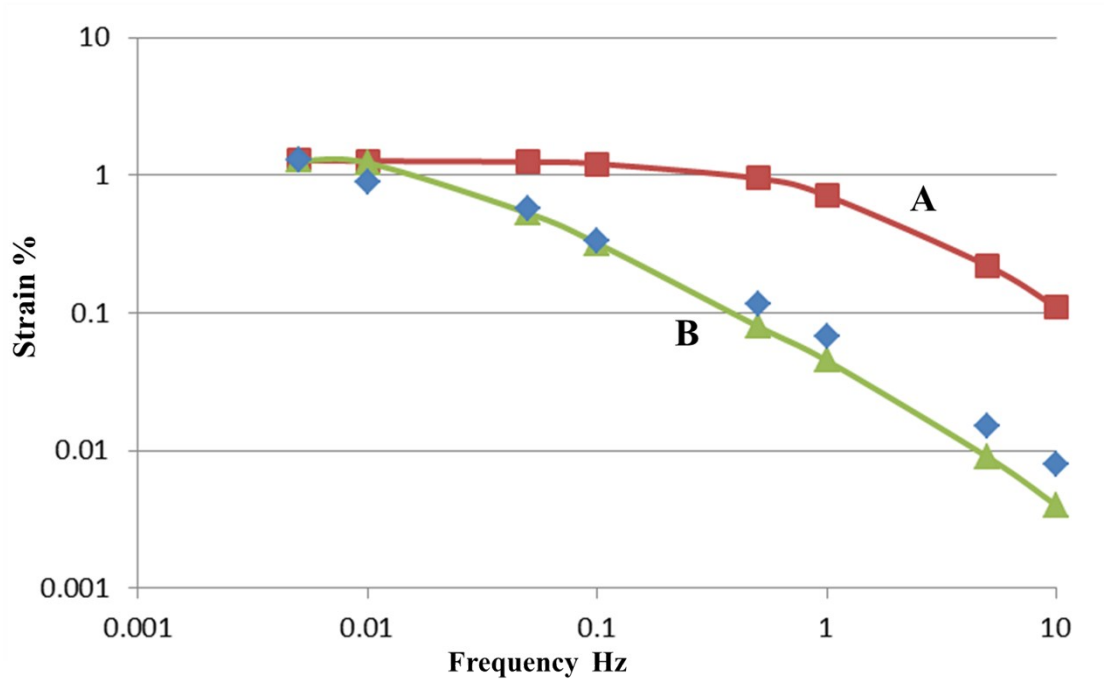


Figure S4 Measured (blue symbols) and simulated (curves) data showing the frequency dependence of the strain for a PEDOT:PSS/VGCF/EMI[CF_3SO_3]/EG 3% device. Curves A and B were calculated using the equivalent circuits in Figures S3(b) and S3(c), respectively.

Table S1. Simulation parameters for the PEDOT:PSS/VGCF/EMI[CF_3SO_3] model that ignores electrode resistance.

IL	C_{VGCF} (F g^{-1})	C (F cm^{-2})	κ (mS cm^{-1}) [a]	R (Ω cm^2)	ε_0 (%)	CR (s)
EG 3%	78.2	0.2874	4.4	0.455	1.28	0.1308
EG 30%	88.4	0.2836	4.4	0.455	1.18	0.1290
EG 0%	76.9	0.2186	4.4	0.455	0.77	0.0994

[a] ref. [S3]

Table S2. Simulation parameters for the PEDOT:PSS/VGCF/EMI[CF_3SO_3] 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)
EG 3%	0.2874	13.5	14.0	4.204
EG 30%	0.2836	14.1	14.6	4.141
EG 0%	0.2186	14.1	14.6	3.192

$\mathbf{R_{el}}$ = area of the electrode film (**cm^2**)/(electrical conductivity (**S cm^{-1}**) × thickness of the electrode film (**cm**)) **(Ref. [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.