Supplementary Materials: Environmental Effects on the Polypyrrole Tri-layer Actuator

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1. Galvanostatic Polymerization of Polypyrrole (PPy)

Constant current (Chronopotentiometry method) is applied between the working electrode (gold sputtered poly(vinylidene fluoride) (PVDF)) and counter electrode (stainless steel mesh) for polymerization of the film. During polymerization, voltage continuously is monitored for 12 h, as shown in Figure S1.

It has been seen that conducting Polypyrrole (PPy) films depends upon suitable polymerization conditions. For instance, appropriate voltage and temperature enhances the electrical conductivity, molecular anisotropy and uniform morphology. PPy synthesis at lower temperatures produces higher electronic conductivity, longer conjugation length, structural order and fewer structural defects. Solvents such as dimethyl formamide (DMF), dimethyl sulphoxide (DMSO), and pyridine impede the anodic electro-polymerization of pyrrole monomer when the pH level is high. Therefore, by considering the effect of the solvent, a slight amount of water (1%) has a substantial influence on the polymerization process and improves the structure and mechanical properties of the film [1].

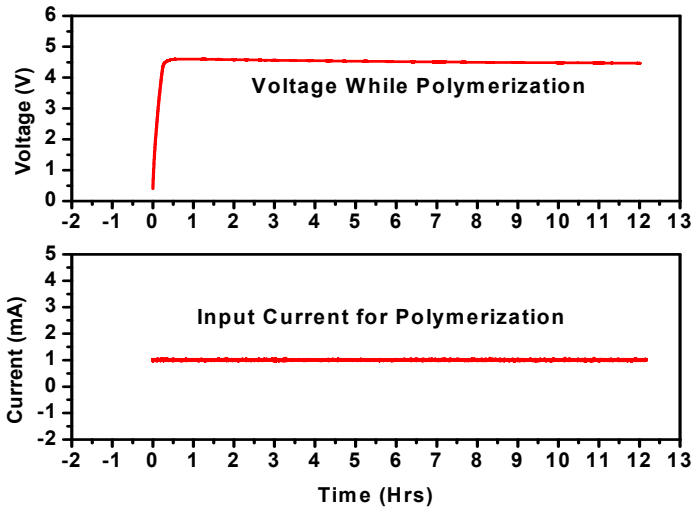

Figure S1. The galvanostatic polymerization of PPy on the gold-coated PVDF film.

2. Quantitative Analysis of Tri-layer Actuator

In this section, the equivalent width method is implemented because of the comparable difference between the moduli of elasticity of the PVDF and PPy [2]. This technique is used to swell the layer with the higher modulus of elasticity, in order to combine the whole structure into a single material with the lower modulus of elasticity. To maintain the same flexural rigidity, the higher modulus of elasticity width is increased by $n = \frac{E_2}{E_1}, E_2 > E_1$. It has been already demonstrated that [2] the moduli of elasticity of PVDF and PPy are approximately 117 MPa and 190 MPa, respectively.

Therefore:

$$n = \frac{E_{PPy}}{E_{PVDF}} = 1.624$$

So, the new cross-section of the PVDF is shown in Figure S2. For commercial PVDF $t_1 = 110$ µm, whereas the thickness of PPy is approximately 8 µm (from the SEM). Therefore, the new equivalent width is $n \times b$ = 1.624 mm (where "b" is normalized to 1 mm). The area moment of inertia is $I = 201.51 \times 10^{-6}$ mm^4 [3].

The bending stress σ is calculated from:

$$\sigma = \frac{Mc}{I}$$

where moment of the inertia I is for new cross-section, "c" is half of the total thickness, which is h_{thick} = 126 µm (110 µm PVDF + 16 µm of PPy), and M is the internal bending moment generated by the electrochemical process. The active layer (PPy) is expected to generate the same bending moment under the same input voltage. Therefore, the bending stress of the actuator is in proportion to the $\sigma = c/I$ ratio. Bending stress in our case is 312.65 for the 110-µm thick PVDF and 8-µm PPy (with an active layer on both sides).

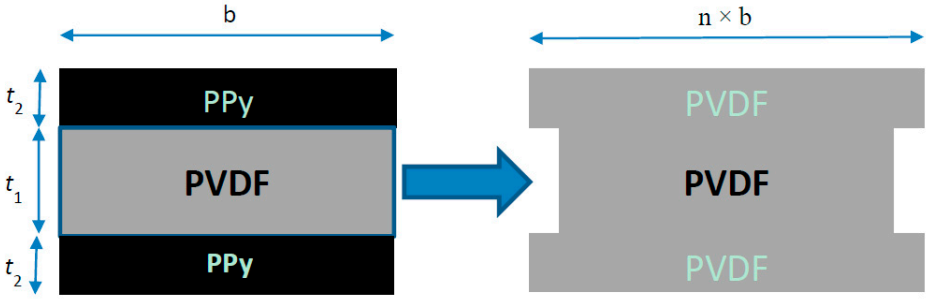

Figure S2. Laminated structure of a polymer and its equivalent cross-section as per equivalent width technique. The mechanical properties of the single material are equivalent to other multi-structure materials.

3. Displacement and Current in Inert/Open Air with Respective Time

Table S1. Displacement and current in inert/open air with respective time.

Time (mins)	Displacement Air (cm)	Current Air (mA)	Displacement Inert (cm)	Current Inert (mA)
0	0	0	0	0
0.5	2.15	39.392	2.375	72.7
1.5	2.24	40.336	2.5	70.3
2.5	2.33	42.523	2.5	68.8
3.5	2.31	43.25	2.5	67.9
4.5	2.39	43.4	2.5	67.4
5.5	2.39	43.826	2.5	67.4
6	2.39	43.826	2.5	67.4

4. PVDF FTIR at Different Position

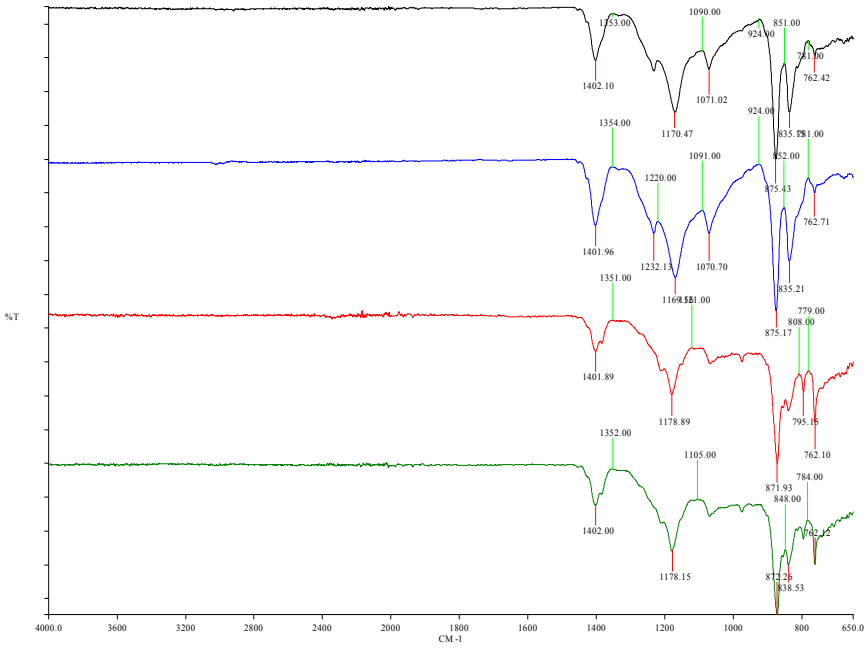

Figure S3. Fourier transform Infrared (FTIR) characteristics of PVDF in four places.

5. Lifetime Test of the PPy Tri-layer Actuator

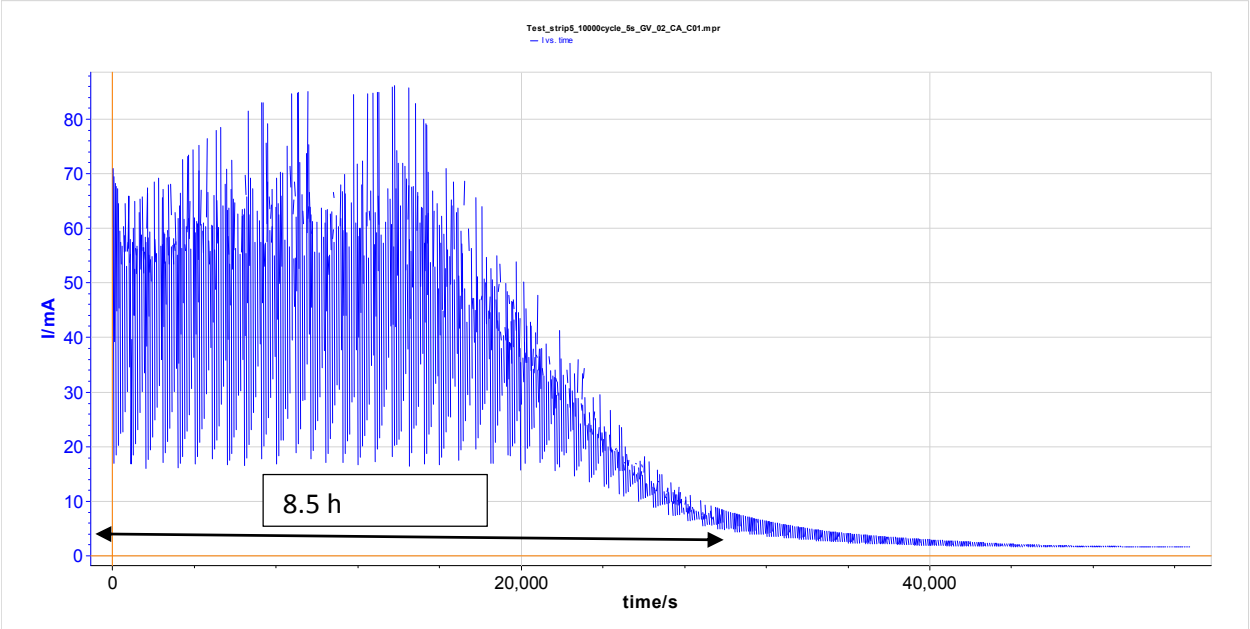

Figure S4. Lifetime test of the PPy tri-layer actuator.

6. Actuator Performance at Different Frequencies

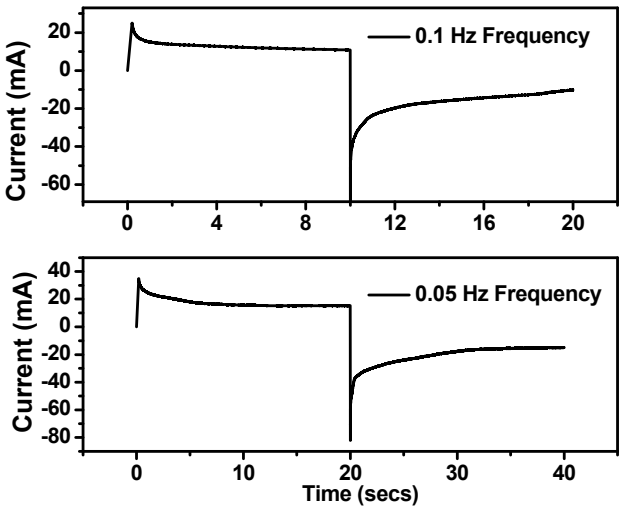

Figure S5. Actuator performance at different frequencies.

References

1. Ansari, R. Polypyrrole conducting electroactive polymers: synthesis and stability studies. *J. Chem.* **2006**, *3*, 186–201.
2. John, S.W.; Alici, G.; Cook, C.D. Validation of resonant frequency model for polypyrrole trilayer actuators. *IEEE/ASME Trans. Mechatron.* **2008**, *13*, 401–409.
3. Gaihre, B.; Alici, G.; Spinks, G. M.; Cairney, J. M. Synthesis and performance evaluation of thin film PPy-PVDF multilayer electroactive polymer actuators. *Sens. Actuators A Phys.* **2011**, *165*, 321–328.