

Supporting Information

**A Highly Transparent Crosslinkable Radical Copolymer  
Thin Film as the Ion Storage Layer in Organic  
Electrochromic Devices**

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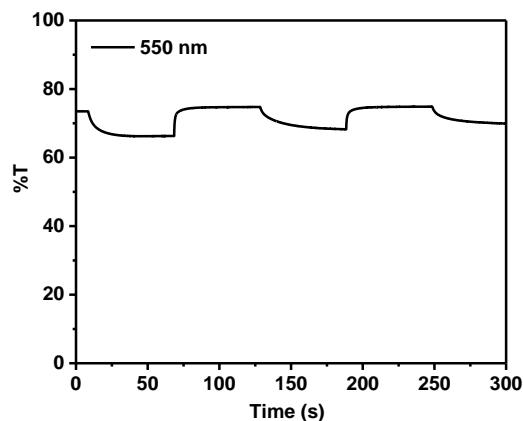
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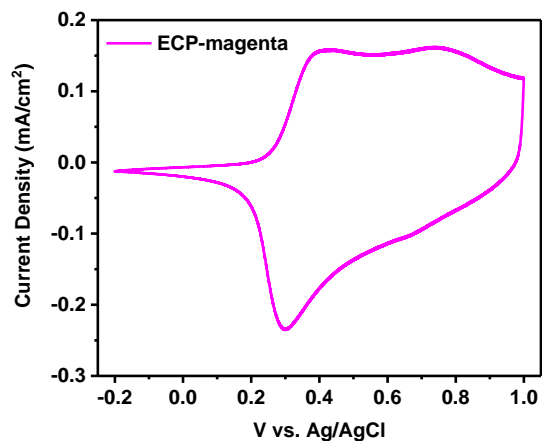
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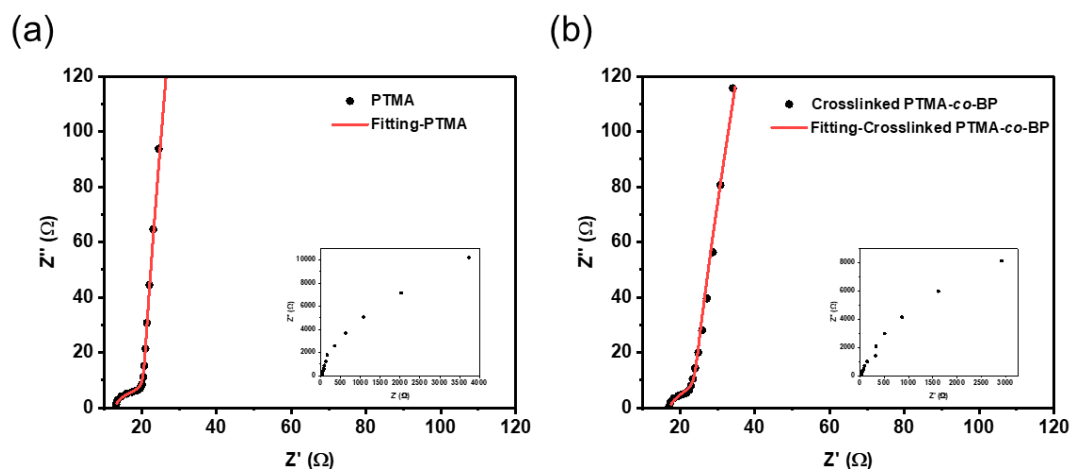


**Figure S1.** Double-potential step chronoamperometry of a two-electrode cell of PTMA-*co*-BP and pre-oxidized ECP-magenta. The voltage was switched between -1.0 V and 2.2 V.



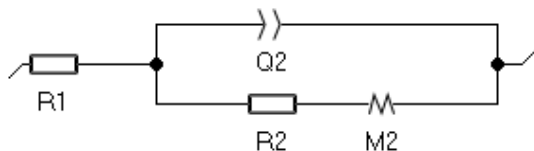
**Figure S2.** Cyclic voltammogram of ECP-magenta with a thickness of  $\sim 240$  nm at the scan rate of  $40 \text{ mV s}^{-1}$ .

The EIS experiments were obtained using a 20 mV perturbation amplitude ( $V_{\text{rms}} = 14.4 \text{ mV}$ ) in a frequency ranging from 1.0 Hz to 1.0 MHz. The stray points with a negative value in high frequency and the deviated points in low frequency were removed before data fitting.



**Figure S3.** (a) EIS measurements of the PTMA thin film (inset: the full spectrum of EIS), and (b) the crosslinked PTMA-co-BP thin film (inset: the full spectrum EIS).

Scheme S1. R(Q(RM)) Equivalent Circuit



Both EIS measurements show small semi-circle in the high-frequency region which is related to the double layer capacitance and charge transfer resistance. In the low-frequency region, the impedance plots show a line approaching  $90^\circ$ . While coming to the lower frequency  $\sim 10$  Hz, they start to deviate and appear like an arc shape. (Figure S1) Thus, a modified Randles circuit with a restricted linear diffusion element was used as the equivalent circuit to model the EIS spectra after removing the deviation points.<sup>1</sup> (Scheme S1) From the fitting results, the  $R_{ct}$  for PTMA and PTMA-co-BP are  $14.2 \Omega$  and  $15.7 \Omega$ , respectively. The similar values of  $R_{ct}$  suggest the crosslinking of PTMA-co-BP thin films did not result in an increase of ionic resistances.

## Reference

- (1) Bobacka, J.; Lewenstam, A.; Ivaska, A. Electrochemical Impedance Spectroscopy of Oxidized poly(3,4-Ethylenedioxythiophene) Film Electrodes in Aqueous Solutions. *J. Electroanal. Chem.* **2000**, *489* (1), 17–27.