# Supporting Information

### Phenalenyl Based Neutral Radical as a Novel Electrochromic Material Modulating Visible to Short-Wave Infrared Light

## Dejan Stekovic\*<sup>a,b</sup>, Prof. Mikhail E. Itkis\*<sup>a,b,c</sup>

<sup>a</sup> Department of Chemistry, University of California, Riverside, California 92521, United States

<sup>b</sup> Center for Nanoscale Science and Engineering, University of California, Riverside, California 92521, United States

<sup>c</sup> Department of Chemical and Environmental Engineering, University of California, Riverside, California 92521, United States

\* E-mail: dstek001@ucr.edu , mitkis@engr.ucr.edu

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#### **Experimental Section**

Materials: Propylene Carbonate and hydroquinone were used as received (Sigma Aldrich).

*Synthesis of [PLY(O,NBu)]*<sub>2</sub>*B*: [PLY(O,NBu)]<sub>2</sub>B was synthesized as the cationic salt ([PLY(O,NBu)]<sub>2</sub>B<sup>+-</sup>BPh<sub>4</sub>) according to the literature.<sup>1</sup>

*Preparation of Electrochromic solution:* 8 mg of  $[PLY(O,NBu)]_2B^+$  <sup>-</sup>BPh<sub>4</sub> (9.0 x 10<sup>-6</sup> mol) along with 2 mg hydroquinone (1.8 x 10<sup>-5</sup> mol) was dissolved in 1 mL of propylene carbonate.

*ITO Sandwich Device preparation:* The electrochromic solution is sandwiched between two ITO coated glass slides (20 Ohm/sq, Thin Film Devices, Inc, Anaheim, CA) and sealed utilizing double sided tape with 3.5 mm x 13 mm rectangle cut aperture as shown in Figure 2a.

*MT-SWNT Sandwich Device preparation:* Similar to our previous report,<sup>2</sup> the thin MT-SWNT films (30 nm thick) were made utilizing vacuum filtration of the dispersion of large diameter (1.2-1.7 nm) 99% separated (IsoNantube-M) metallic SWNTs purchased from Nanointegris Inc. The films were transferred onto glass substrates bridging a 2 mm gap between predeposited Ti(15nm/Pt(150 nm) electrodes. On one substrate, an adhesive seal frame (0.25 mm thick) (Frame-SealTM, Bio-Rad Laboratories) is placed. The cell is filled with electrochromic solution and another matching MT-SWNT on glass substrate is placed overtop (see picture in Figure S4).

*Spectroscopy/Electrochromic Measurements:* Transmittance spectra were recorded on a Carry 5000 UV-Vis Spectrophotometer (Agilent Technology). The potential was applied using a model DS345 synthesized function generator (Stanford Research Systems).

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**Fig. S1:** (a) Transmittance and (c) absorbance spectra of device, glass and ITO on glass in the range of interest (400-1000 nm). (b)Transmittance and (d) absorbance spectra of glass and ITO on glass showing a high absorption of ITO above 1200 nm.



**Fig. S2:** (a) Transmittance and (b) absorbance spectra of [PLY(O,NBu)]<sub>2</sub>B based device components using MT-SWNTs as electrodes. The MT-SWNTs absorb very little SWIR light while propylene carbonate absorbs a significant amount.



0V

2V

**Fig. S3:** Pictures of the MT-SWNT electrode devices in their (left) transmissive and (right) black states.

## **References:**

- 1. X. Chi, M. E. Itkis, K. Kirschbaum, A. A. Pinkerton, R. T. Oakley, A. W. Cordes and R. C. Haddon, *J. Am. Chem. Soc.*, 2001, **123**, 4041-4048.
- 2. D. Stekovic, B. Arkook, G. Li, W. Li, E. Bekyarova and M. E. Itkis, *Adv. Mater. Interfaces*, 2018, 1800861.