

Exploring unbalanced electrode configurations for electrochromic devices

Sofiane Hassab¹, D. Eric Shen², Anna M. Österholm², John R. Reynolds², Javier Padilla¹

¹Department of Applied Physics, ETSII, Technical University of Cartagena (UPCT), Cartagena, 30202, Spain.

²School of Chemistry and Biochemistry, School of Materials Science and Engineering, Center for Organic Photonics and Electronics (COPE), Georgia Institute of Technology, Atlanta, GA 30332-0400, United States.

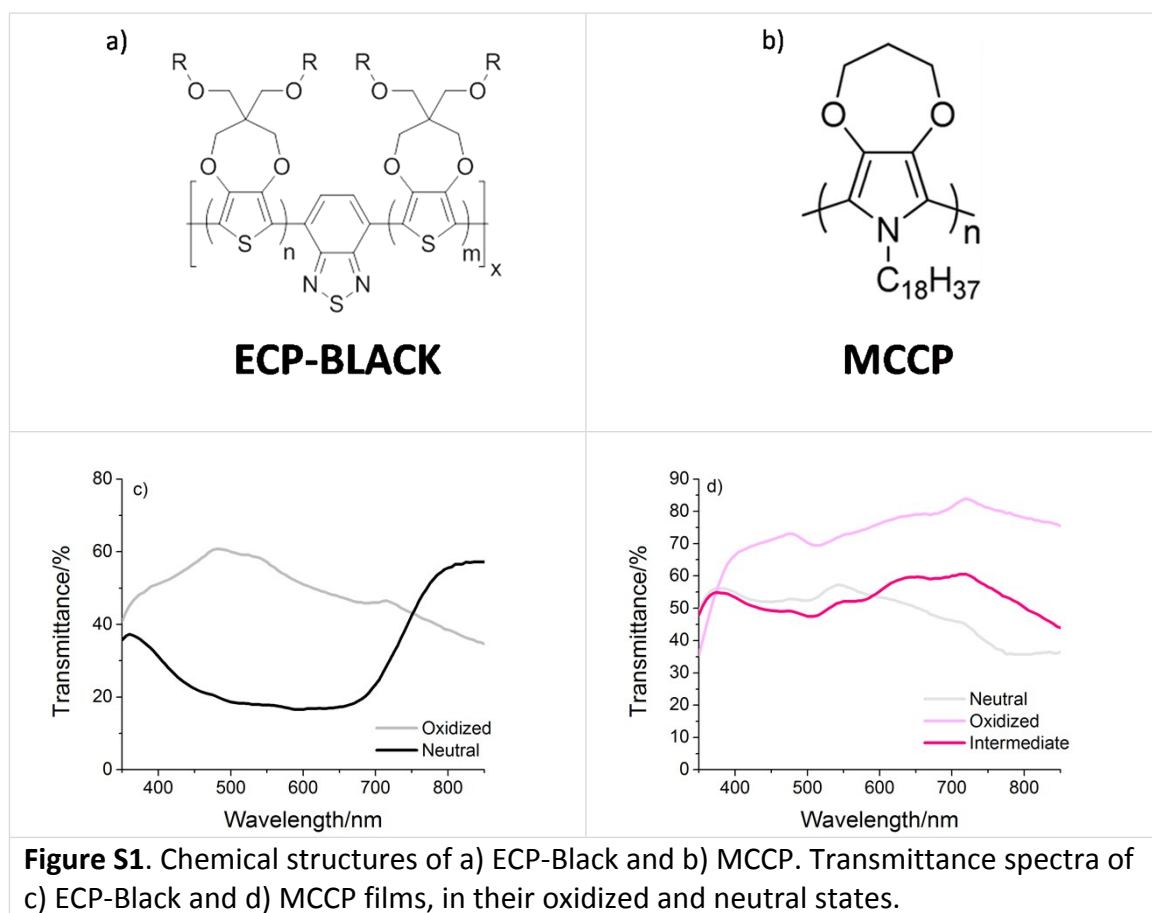


Table S1. Properties of ECP-Black and M CCP

	Optical bandgap / eV	Coloration efficiency / cm^2C^{-1}	Oxidation potential / V vs. Ag wire
ECP-Black ^{1,2}	1.62	110 (@500 nm)	0.57
M CCP ³	2.96	35 (@555 nm)	0.39

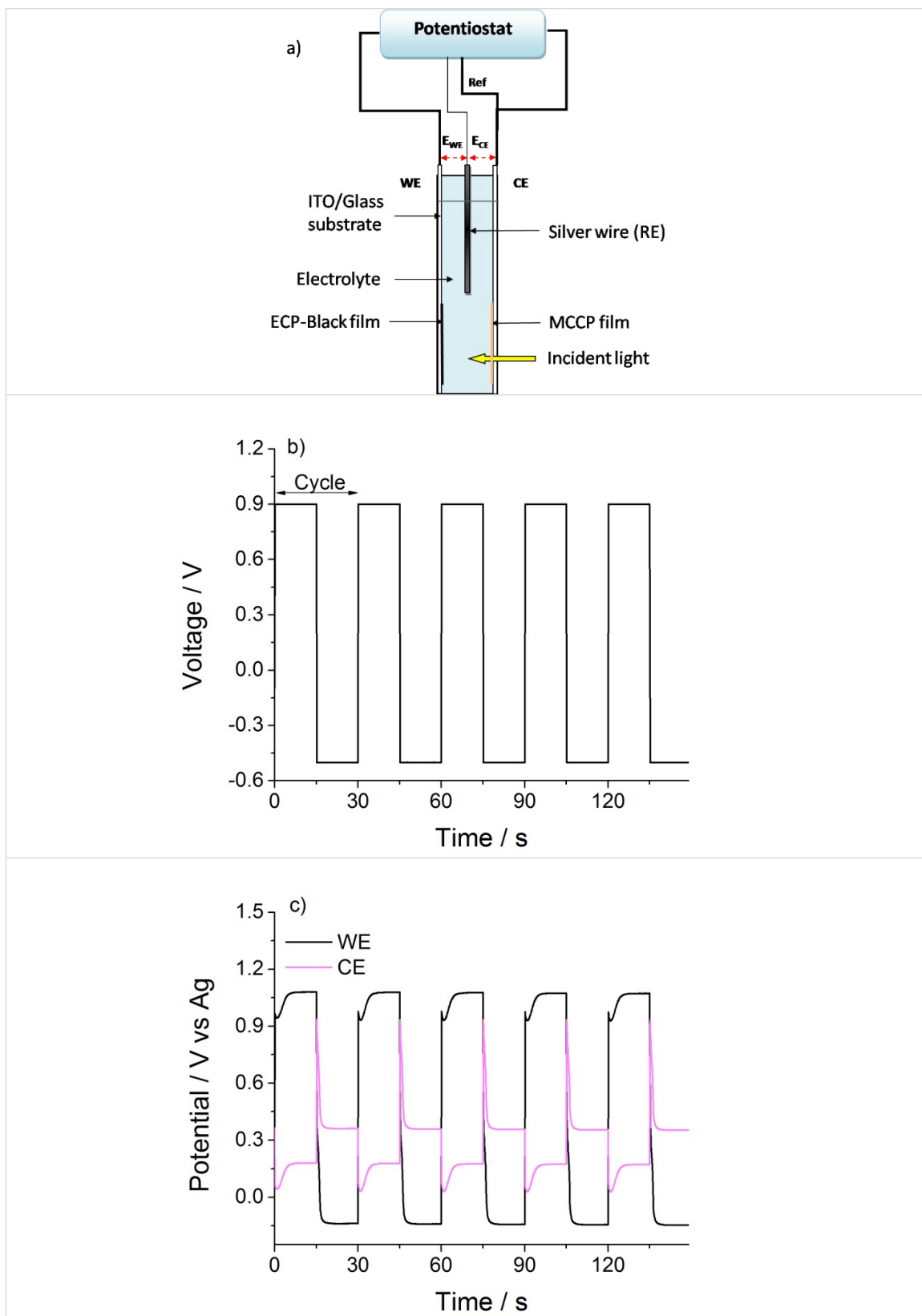


Figure S2. a) Setup of ECP-Black/MCCP electrochromic cells in two-electrode configuration, with an additional reference electrode included to monitor individual potentials of each film (E_{we} and E_{ce} are, respectively, potentials of ECP-Black and MCCP relative to silver wire RE). b) Applied cell voltage as a function of time and c) an example of simultaneously recorded potentials vs. RE obtained independently for

each electrode, for a device constructed with ECP-Black having a charge to switch of 4.7 mC/cm^2 and MCCP having a charge to switch of 9.7 mC/cm^2 .

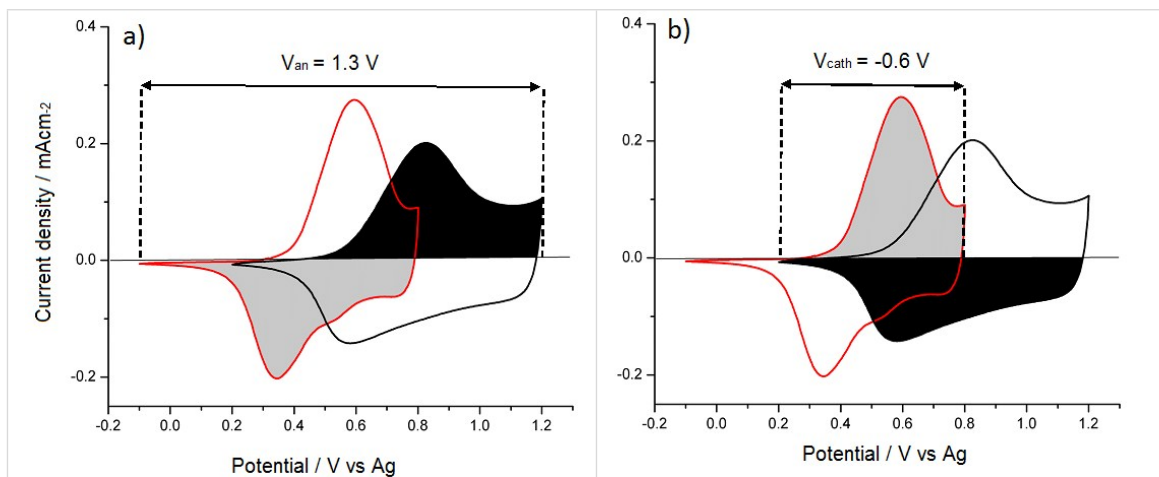


Figure S3. Voltammograms of ECP-Black and MCCP films, in a balanced configuration, where full switching of both materials is obtained when operating the cell. a) Anodic cell voltage is determined from the difference between the potential of an oxidized ECP-Black (1.2 V vs. Ag), and the potential of a neutral MCCP (-0.1 V vs. Ag), that is 1.3 V. b) Cathodic cell voltage is determined from the difference between the potential of a neutral ECP-Black (0.2 V vs. Ag), and the potential of an oxidized MCCP (0.8 V vs. Ag), that is -0.6 V. Total cell voltage necessary is therefore 1.9 V.

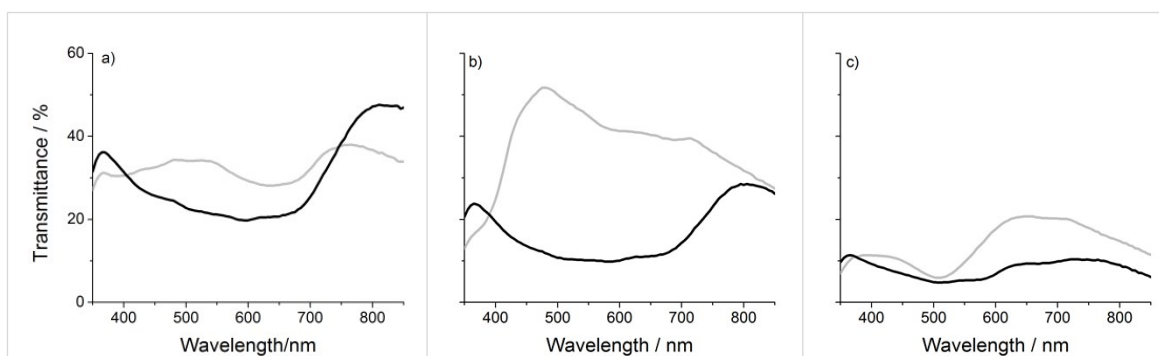


Figure S4. Transmittance spectra of cells with a) 0.2, b) 0.9 and c) 2.9 ratio.

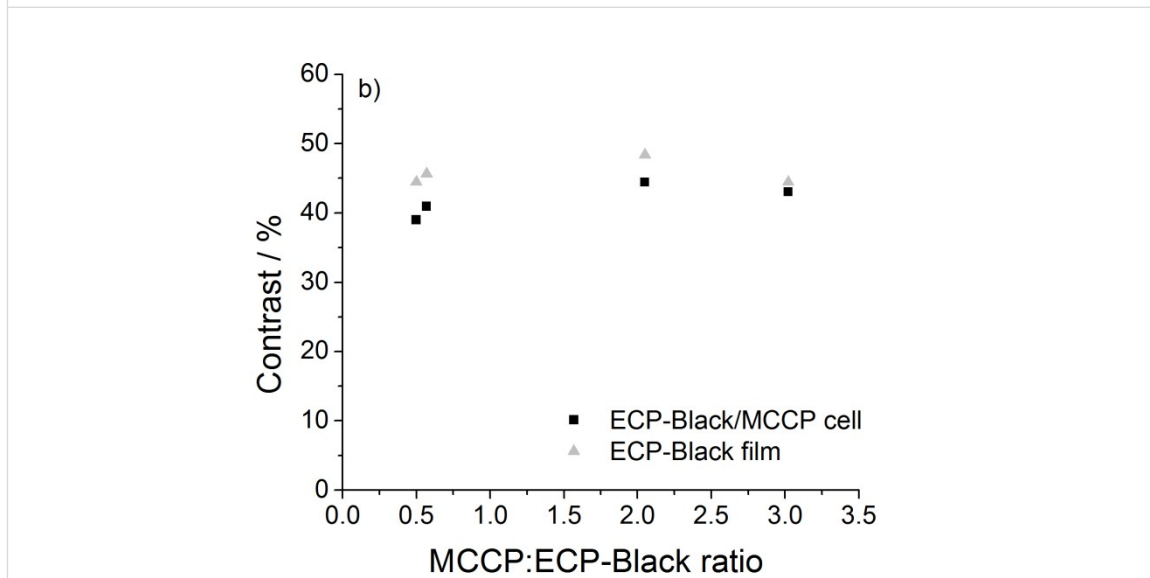
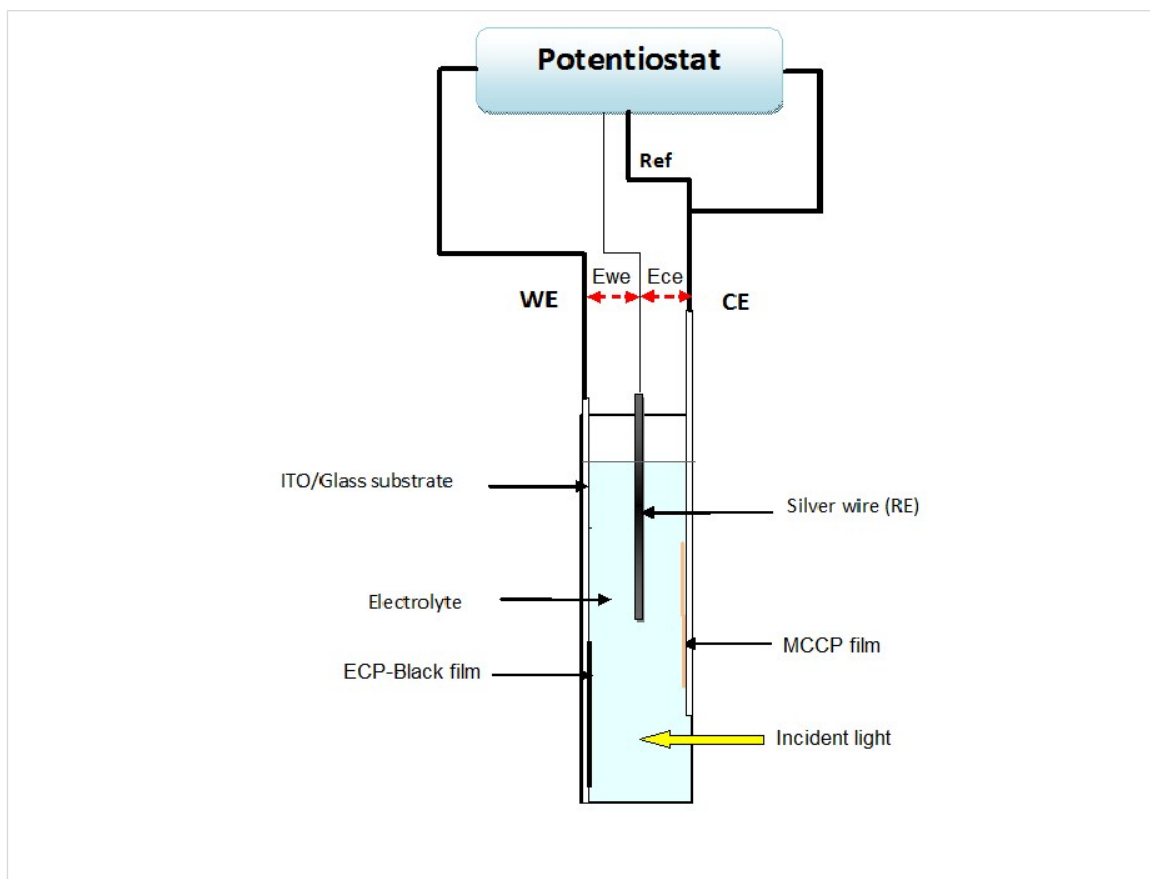


Figure S5. a) Set-up of Black-ECP/MCCP electrochromic cells in a two-electrode configuration, with an additional reference electrode included to monitor individual potentials of each film, and removing the optical contribution of the MCCP film (E_{we} and E_{ce} are, respectively, potentials of Black and MCCP relative to silver wire). b) Contrast obtained with cells for which MCCP films do not contribute optically (black squares) and contrast of the corresponding black film alone (grey triangles).

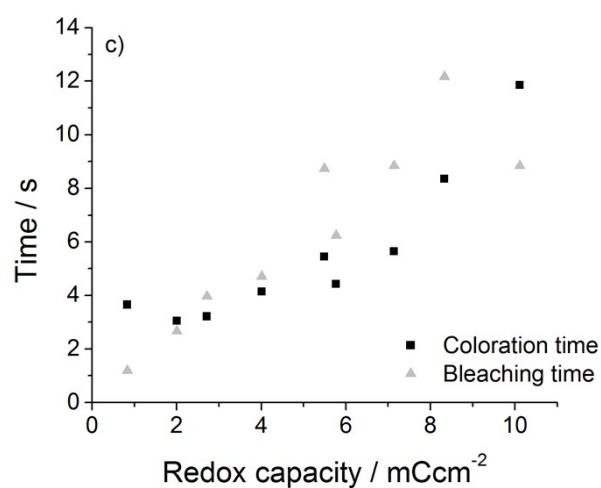
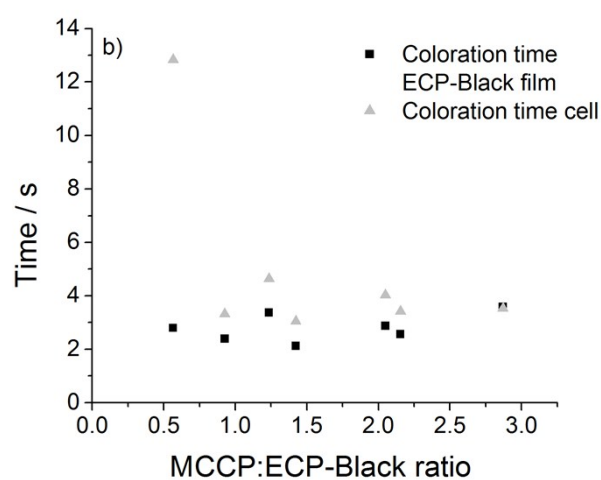
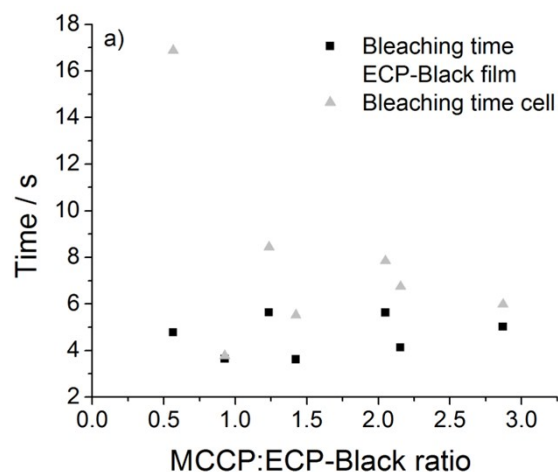


Figure S6. a) Bleaching and b) coloration times of cells with different redox capacity ratios, as well as the corresponding times for an ECP-Black film in a three-electrode cell c) Bleaching and coloration times of MCCP films as a function of the redox capacity

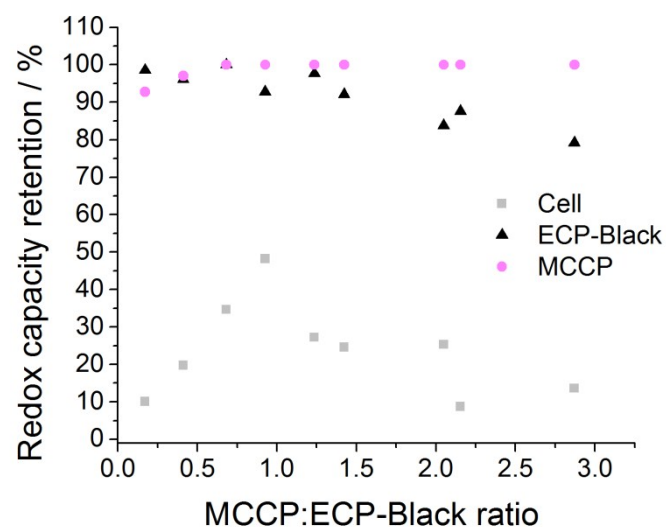


Figure S7. Redox capacity retention after 2500 switches. Values obtained from cyclic voltammograms taken before and after the 2500 cycles test

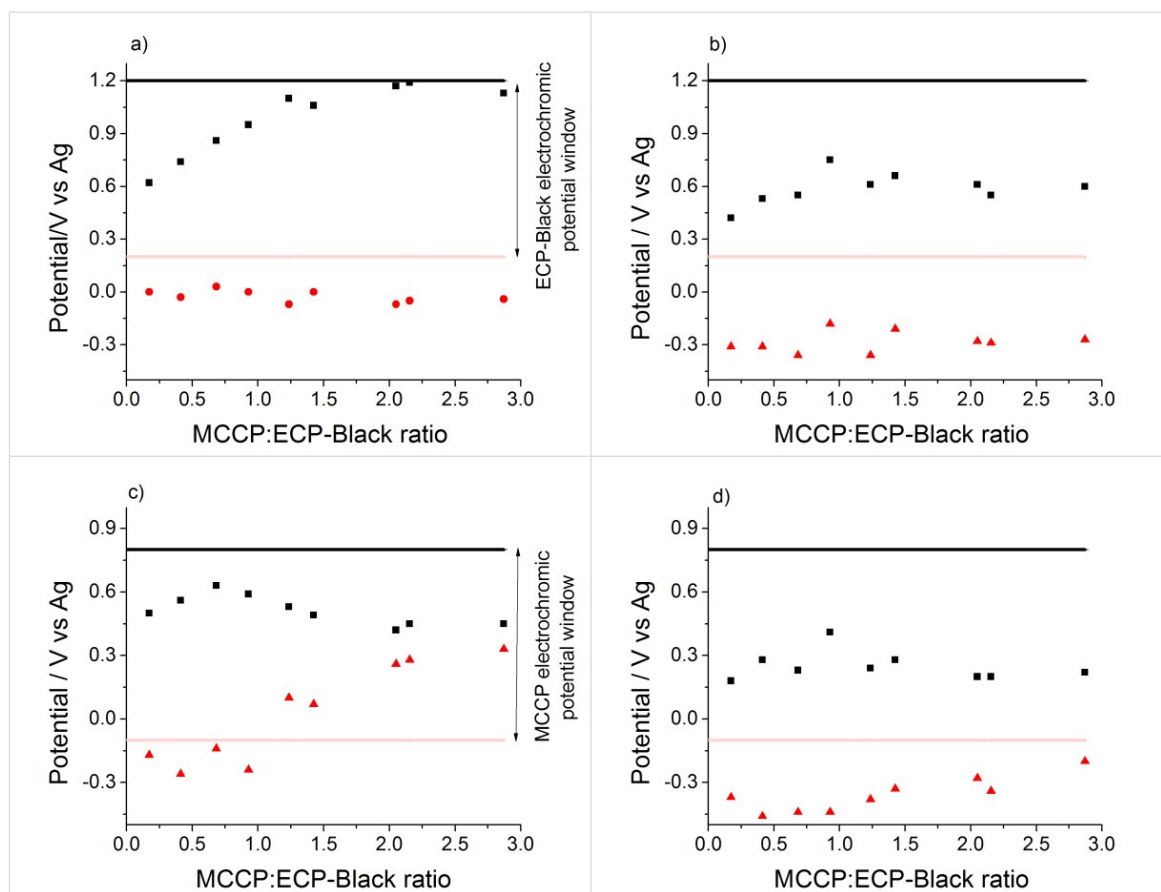


Figure S8. Potential evolution for ECP-Black electrodes as a function of redox capacity ratio in the cell a) before and b) after 2500 switches. Potential evolution for MCCP electrode as a function of redox capacity ratio in the cell c) before and d) after 2500 switches. The electrochromic potential window of the respective films is included as a visual reference (pink and black lines) in the figures.

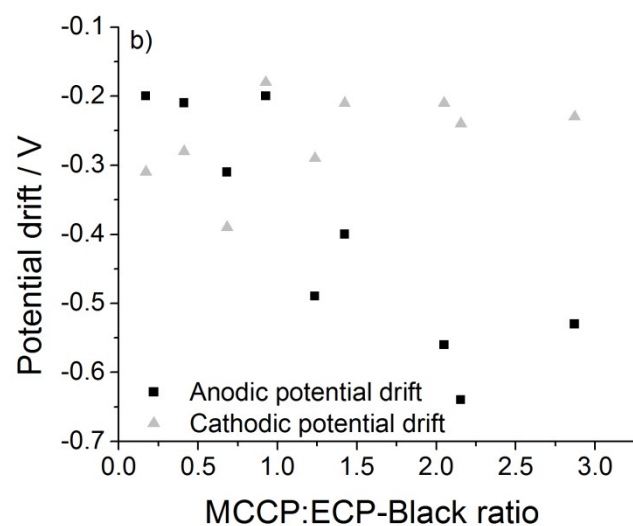
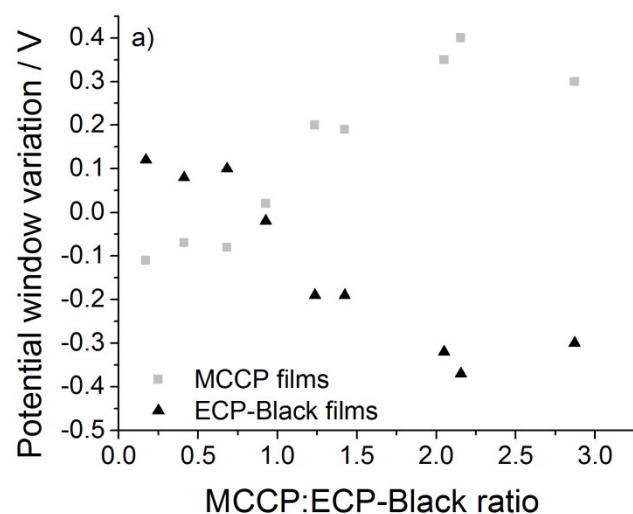


Figure S9. a) Potential window variation of each film during a 2500 cycle test, as a function of the cell redox capacity ratio. The potential window of each polymer is the difference between the anodic and cathodic potentials reached, and the variation of this potential window during the test is calculated from the difference between the final and initial cycles.

b) Anodic and cathodic potential drifts for ECP-Black films as a function of cell redox capacity ratio. Anodic and cathodic potential drifts are defined as the difference between the potential reached in the last cycle and that reached in the first cycle.

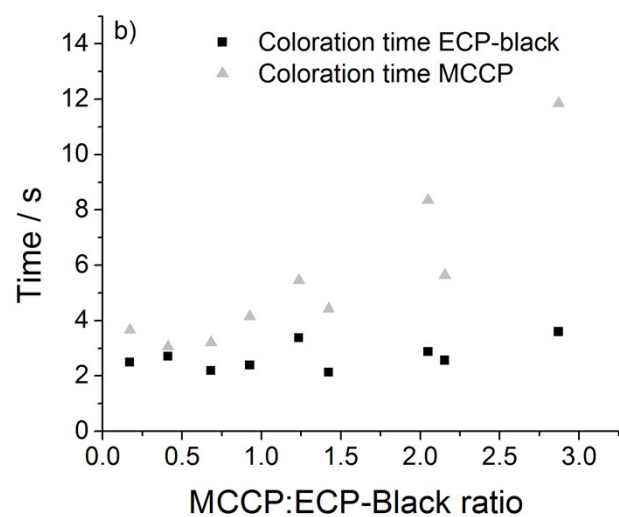
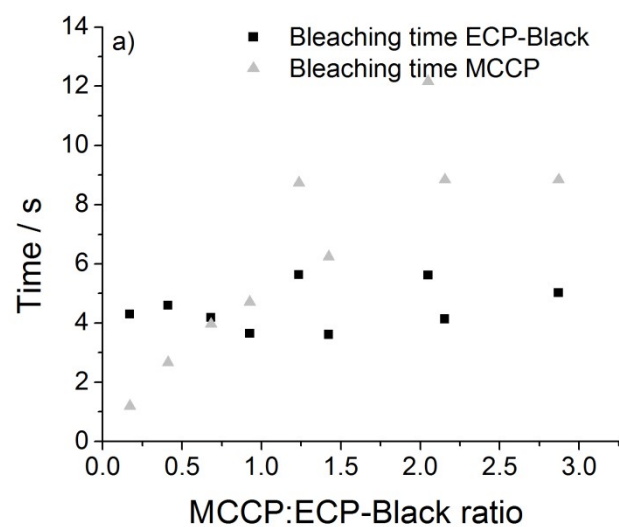


Figure S10. a) Bleaching times and b) coloration times for ECP-Black and M CCP films used for the different cells.

References

1. P. Shi et al., *Adv.Mater.*, 2010, **22**, 4949-4953.

2. J. Padilla, A. M. Osterholm, A. L. Dyer, J. R. Reynolds, *Sol.Energ.Mat.Sol.C.*, 2015, **140**, 54-60.
3. E. P. Knott et al., *J.Mater.Chem.*, 2012, **22**, 4953-4962.