Supporting information

Single sheet iron oxide based films: Electrochemical properties with *in-situ* UV-vis measurement

Li-Zhi Huang, $*^a$ Tue Hassenkam, ^b Hans Christian B. Hansen, ^a and Morten Jannik Bjerrum^b

^{*a*} Department of Plant and Environmental Sciences, University of Copenhagen, Thorvaldsensvej 40, DK–1871 Frederiksberg C, Denmark.

^b Nano-Science Center, University of Copenhagen, Universitetsparken 5, 2100 København Ø, Denmark.

^c Department of Chemistry, University of Copenhagen, Universitetsparken 5, DK-2100 København Ø, Denmark.



Fig. S 1. AFM image of PDDA-precoated mica surface. The surface is very flat and homogenous slightly different from the original mica surface with a roughness around 2 Å. This roughness reflects the height variation stemming from the PDDA coating and it is negligible compared to that of the subsequent SSI coating (~14 nm in average).



Fig. S 2. Height AFM image of the first layer of PDDA/SSI film deposited on mica surface. The red line in (a) marks the position and length the cross section displayed in (b). The 1-nm step increment shows a single sheet iron oxide (8×30 nm in diameter) lying on top of a stack of single sheet iron oxide.



Fig. S 3. Photography of a bare quartz ITO electrode (left), and a quartz ITO electrode deposited with (PDDA/SSI)₁₄ films (right).



Fig. S 4. Effect of cathodic switching potential (E_c , from -0.5 to -0.9 V) on the UV-vis spectrum of (PDDA/SSI)₂ films on ITO electrode at pH 7 (UV-vis spectrum corresponding to Fig. 5 in the paper). There was no decrease in UV-vis absorbance of (PDDA/SSI)₂ films with E_c at -0.5 V, whereas a decrease was observed with E_c at -0.9 V. The absorbance of bare ITO electrode was subtracted.



Fig. S 5. Stability of (PDDA/SSI)_n films with $E_c = -0.7$ V at pH 12. (ITO electrode deposited with (PDDA/SSI)₂ films, sweep rate: 0.1 V s⁻¹, 0.01 M NaOH electrolyte, blank results from bare ITO electrode under the same conditions).



Fig. S 6. UV-vis absorption spectra for (PDDA/SSI)₁₀ films on ITO quartz electrode (a) at various potentials. The baseline is the absorbance of the (PDDA/SSI)₁₀ films at +0.6 V, and (b) the corresponding absorbance at 341 nm vs. applied potential. (the spectra at +0.5, +0.4, +0.3, +0.2, +0.1, 0 V are not shown due to overlap). In order to further reveal the influence of external potential on the spectrum of $(PDDA/SSI)_n$ films, the working electrode was kept at various cathodic potentials and the spectra were taken when the current reached zero. The mixed anodic and cathodic electrochromism was observed, which is the same in the as spectro-chronoamperometric study. No substantial spectral change was observed if the potential was positive which is in agreement with the observation in the main paper. A substantial decrease of the absorbance in UV region was observed when the potential was lower than -0.4 V which is the potential that cathodic current starts to increase in CV scans.