Supplementary Material (ESI) for *Nanoscale*
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Electronic Supplementary Information (ESI) Available

Photoelectrochemical behaviour of anatase nanoporous films: effect of the nanoparticle organization

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1. Typical spectral irradiance of a 150 W Xe arc lamp

The data can be found at the Newport webpage (http://www.newport.com), concretely at: http://search.newport.com/?q=*&x2=sku&q2=6253. It corresponds with the Xe short arc bulb fabricated by Osram, model XBO 150W/1.

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2. Transmission Electron Microscopy Images of wire-like nanoparticle aggregates

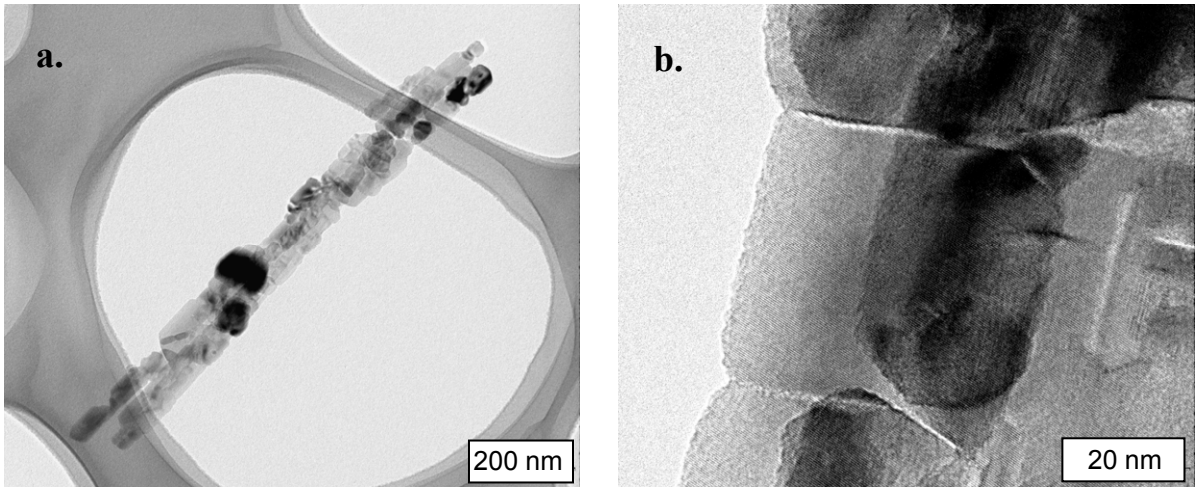

Figure S1: Transmission Electron Microscopy image of a wire-like nanoparticle aggregate (a). Magnified detail of intergrain boundaries (b). The images were obtained from the as-synthesized nanopowder.

3. Electrode Raman spectra

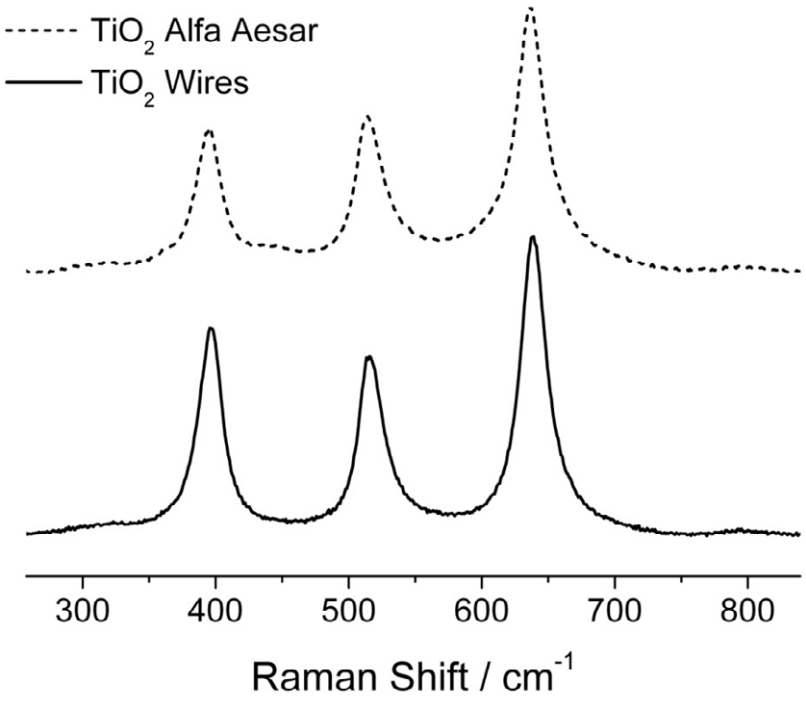

Figure S2. Raman spectra of TiO_2 wire-like nanocrystal aggregates and commercial TiO_2 nanoparticle (Alfa Aesar) thin films deposited on conducting glass.

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4. XRD patterns of the anatase thin films

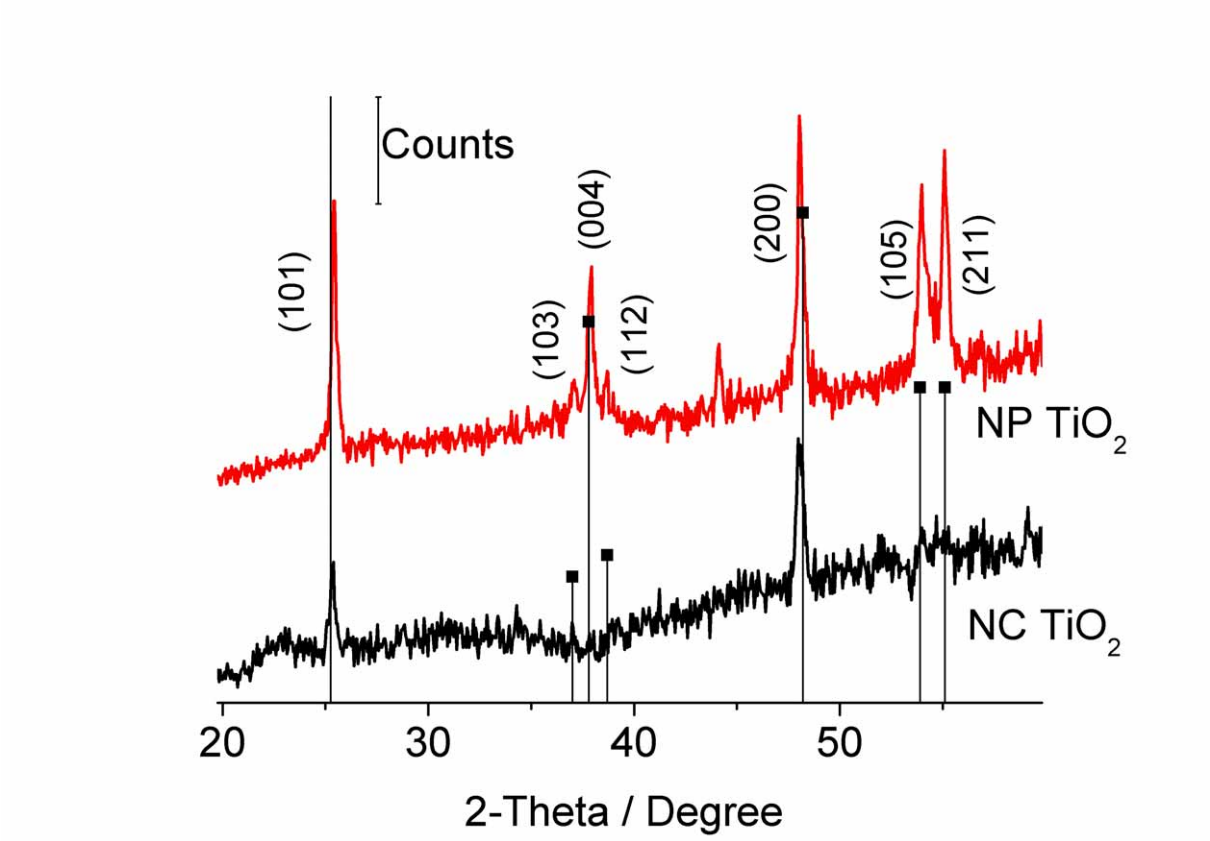

Figure S3. XRD patterns of a nanoparticulate (NP TiO_2) and a nanocolumnar (NC TiO_2) anatase thin film deposited on a fused silica substrate. The smaller peak intensity for the NC TiO_2 layer can be attributed to a smaller amount of material present in the film. A more intense diffractogram for this sample can be found in reference 22.

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5. Cyclic voltammograms obtained at different scan rates

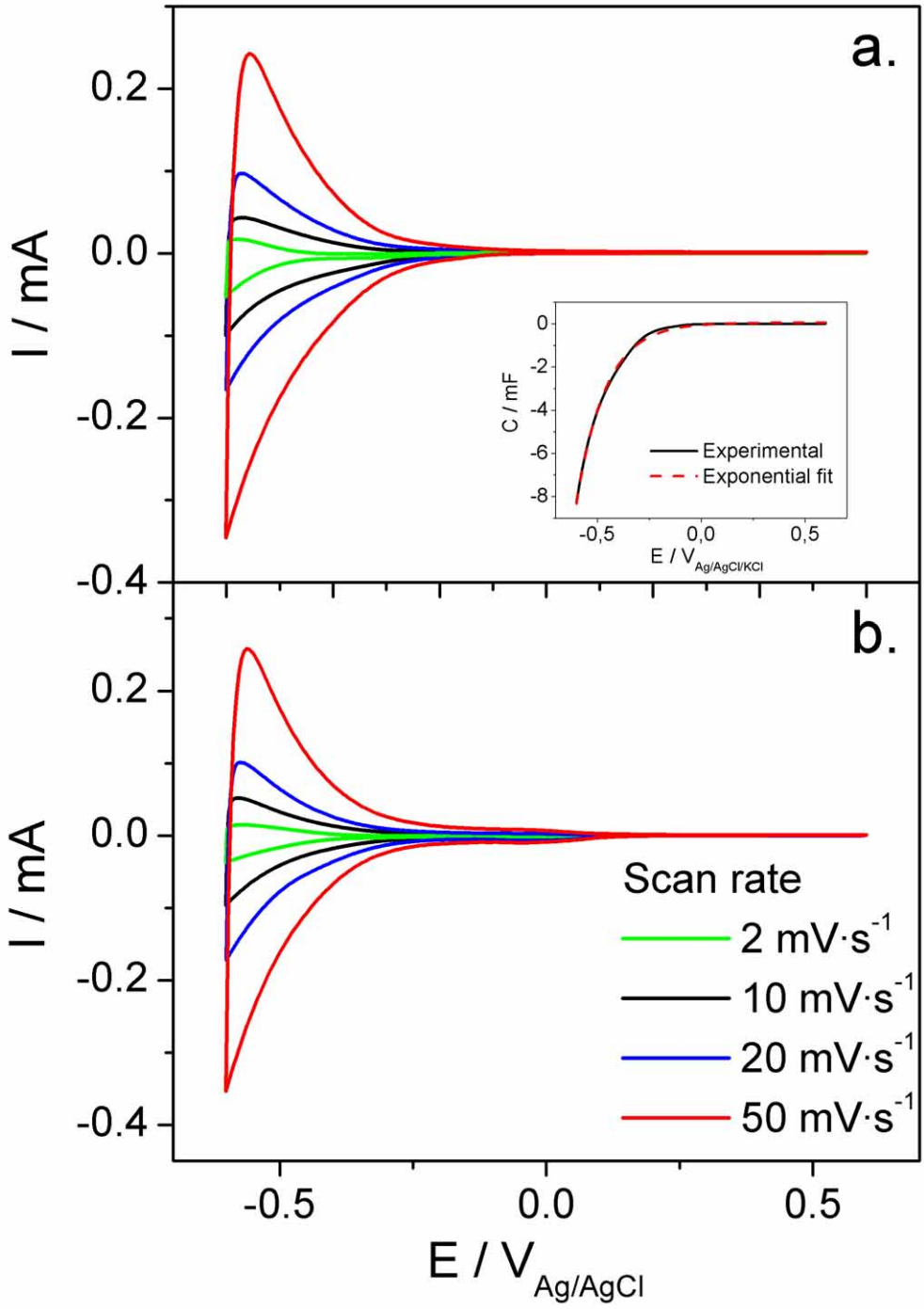

Figure S4. Cyclic voltammetry of a NC electrode (~5 μm) (a) and a NP electrode (~0.7 μm) (b) in the dark at different scan rates. Electrolyte: nitrogen purged 0.1 M $HClO_4$. The inset of Figure a. shows the dependence of the capacitance on the applied potential for the NC electrode.

6. Cyclic voltammograms obtained for randomly distributed anatase nanoparticles after different thermal treatments

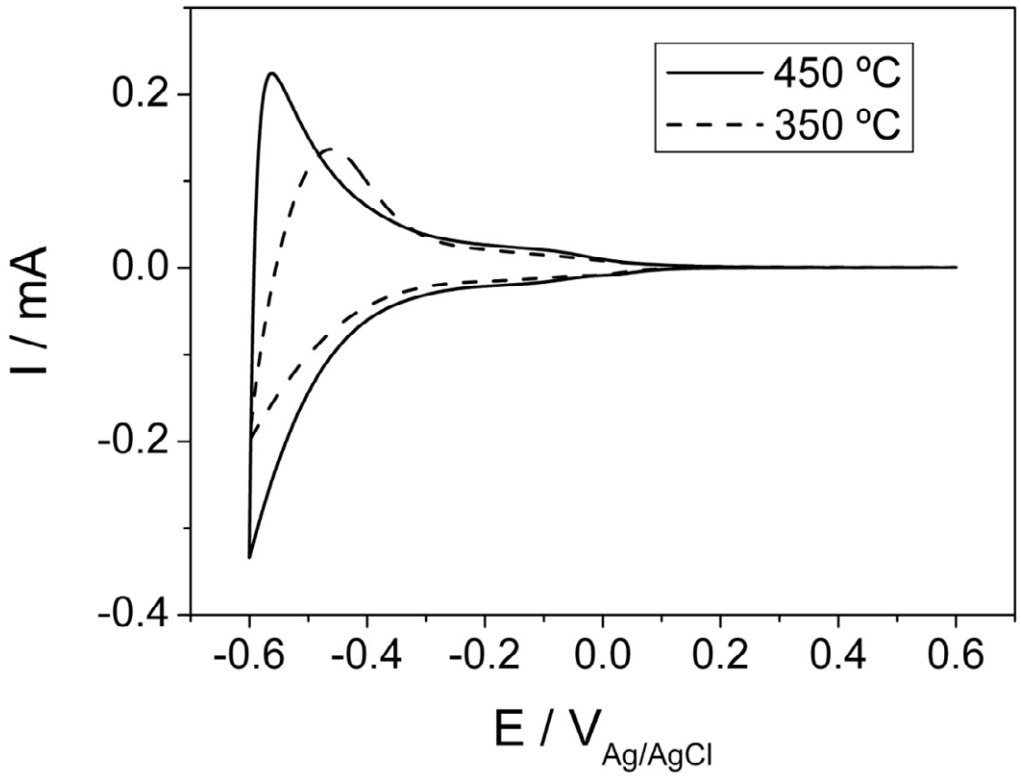

Figure S5. Cyclic voltammograms for a NP electrode in the dark after thermal treatment at either 350ºC or 450ºC. Electrolyte: nitrogen purged 0.1 M $HClO_4$, scan rate: 20 $mV \cdot s^{-1}$. Electrode area ~2 cm^2.

7. Cyclic voltammograms in 0.1 M $HClO_4$ under illumination, in the presence and in the absence of oxygen for randomly distributed nanoparticles and wire-like nanoparticle aggregates

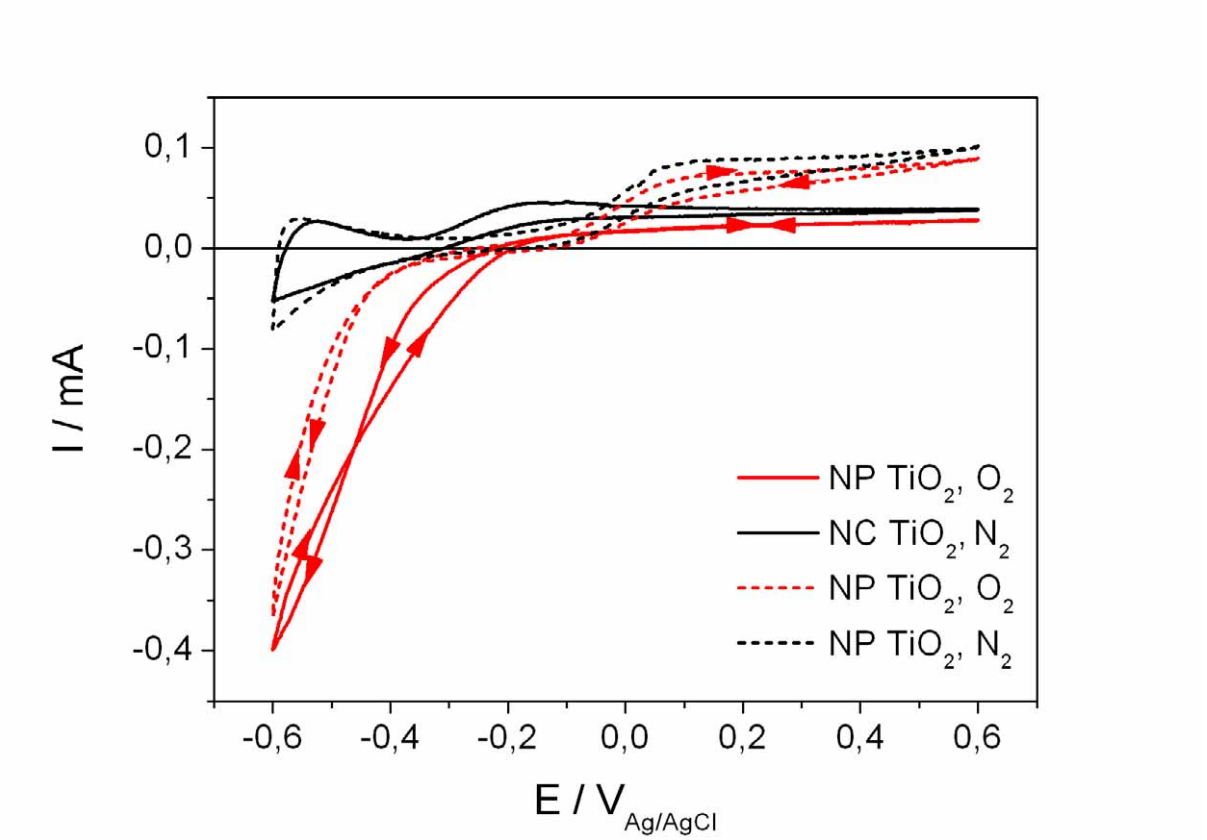

Figure S6. Cyclic voltammetry of a wire-like nanocrystal aggregate electrode (~ 6 µm) and a randomly distributed nanoparticulate electrode (~ 1 µm) (Alfa Aesar) under UV-visible polychromatic illumination (660 $mW \cdot cm^{-2}$). Electrolyte: oxygen (red) or nitrogen (black) purged 0.1 M $HClO_4$; Scan rate: 20 $mV \cdot s^{-1}$; The experiments were performed using a compact TiO_2 blocking layer.

8. Modified Kubelka-Munk function and modified Incident Photon to Current Efficiency (IPCE) versus light intensity

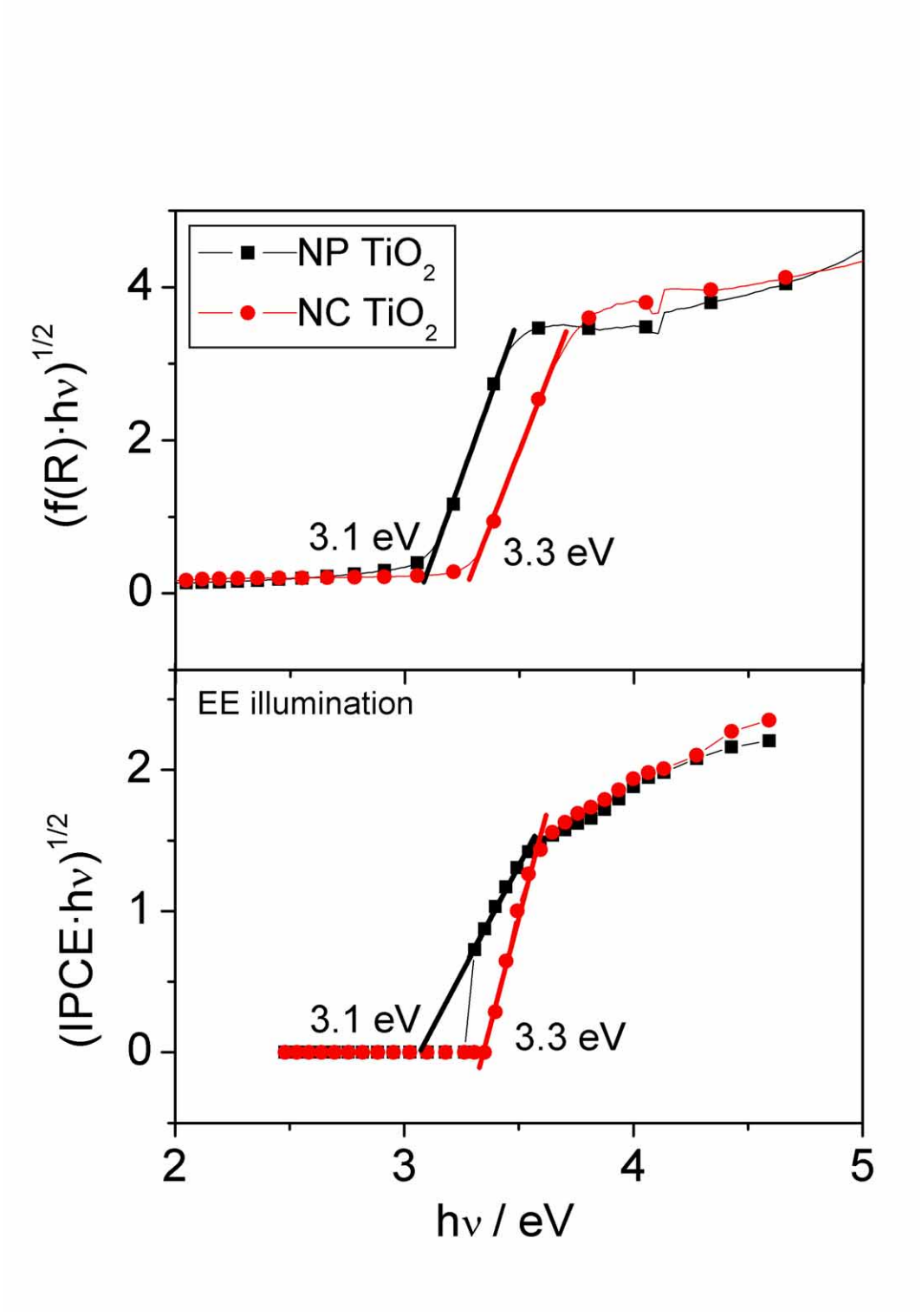

Figure S7. (IPCE · hν)$^{1/2}$ and (transformed reflectance according to Kubelka Munk equation (F(R)) · hν)$^{1/2}$ as a function of hν (photon energy). The IPCE was measured in the presence of formic acid at 0.6 $V_{Ag/AgCl}$.

9. Cyclic voltammograms in 0.1 M $HClO_4$ in the dark and under illumination for NP and NC electrodes with similar film thickness

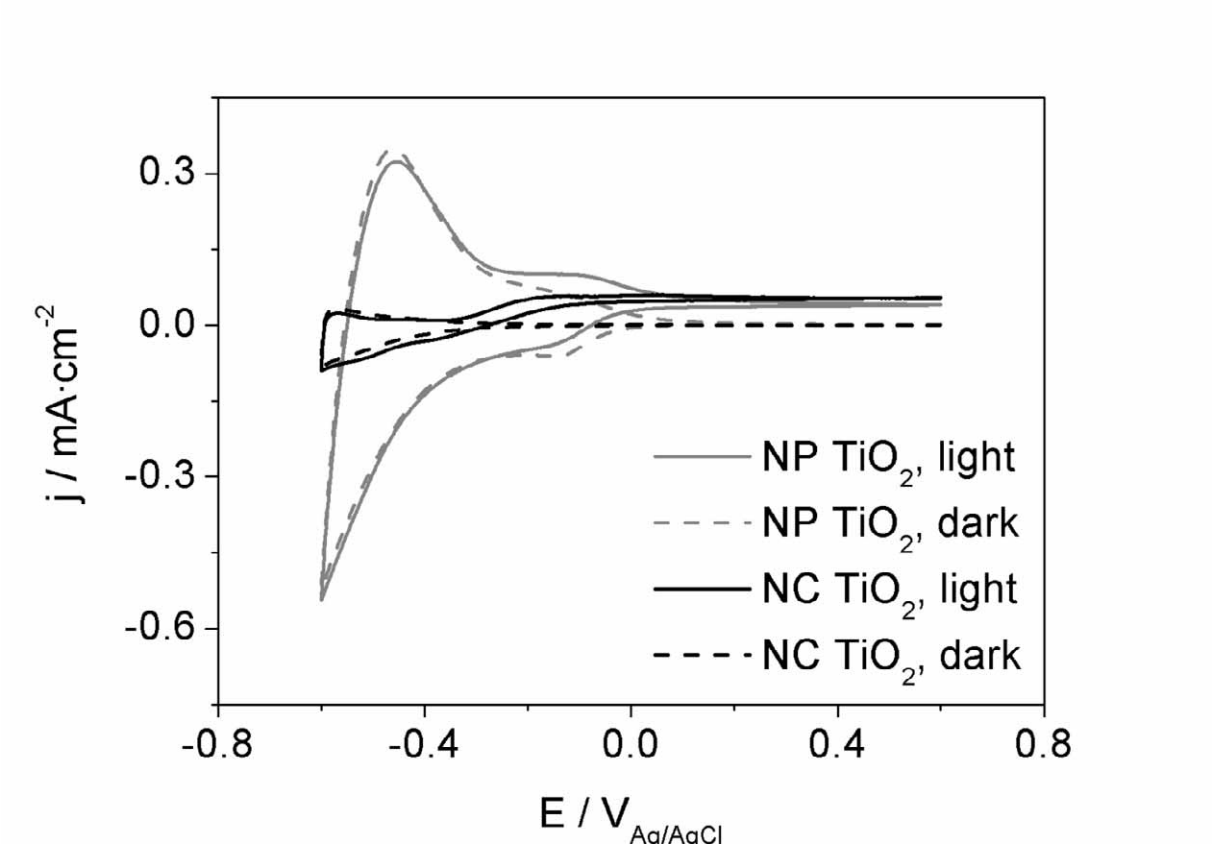

Figure S8. Cyclic voltammograms in 0.1 M $HClO_4$ in the dark. Scan rate: 20 $mV \cdot s^{-1}$