

Electrodeposition of Fe-Complexes on Oxide Surfaces for Efficient OER Catalysis

Sahir M. Al-Zuraiji^{1,2}, Tímea Benkó², Krisztina Frey², Zsolt Kerner² and József S. Pap^{2,*}

¹ Doctoral School on Materials Sciences and Technologies, Óbuda University, H-1034 Bécsi Street 96/b, 1034 Budapest, Hungary; sahir.aziz@energia.mta.hu

² Surface Chemistry and Catalysis Department, Centre for Energy Research, H-1121, Konkoly-Thege Street 29-33, 1525 Budapest, Hungary; pap.jozsef@ek-cer.hu

* Correspondence: pap.jozsef@ek-cer.hu; Tel.: +36-1-392-2222/3284 (J.S.P.)

Contents

Figure S1. (a) CV scans of 1 (0.4 mM) in DCM with added H ₂ O (3 mmol in 0.5 mL acetone) on a BDD electrode (0.071 cm ²); (b) CV scans of 2 (0.4 mM) under the same conditions. Settings: under Ar, ν = 100 mV/s, Pt, and Ag ⁺ /Ag (0.01 M AgNO ₃ , 0.1 M TBAP/acetonitrile), as counter, and reference electrodes, respectively. Red curves: 20 cycles in a broader potential range in both cases show that the anodic oxidation above +1.2 V <i>vs.</i> Fc ⁺ /Fc is required for electrodeposition (compare to the blue CVs, 10 cycles, in a narrower potential range). The green CV was recorded in DCM (10 mL) with added H ₂ O (3 mmol in 0.5 mL acetone) on a BDD electrode (0.071 cm ²) for comparison.	2
Figure S2. Comparison of two parallel electrodeposition experiments using complex 1 (0.8 mM) on ITO (1.5 cm ²) in DCM with added water (3 mmol in 0.5 mL acetone) illustrating good reproducibility. Settings: under Ar, ν =100 mV/s, 20 cycles, Pt, and Ag ⁺ /Ag (0.01 M AgNO ₃ , 0.1 M TBAP/acetonitrile), as counter, and reference electrodes, respectively. See the analogous experiments on FTO in Fig. 3c for comparison.	3
Figure S3. (a) SEM image of 1-ED@FTO as-prepared and cleansed with MilliQ water to remove salt residues, (b) EDX spectrum of the area within the white circle; (c-d) SEM images at different magnifications (see the footers for experimental settings).	4
Figure S4. (a) SEM image of 2-ED@FTO as-prepared and cleansed with MilliQ water to remove salt residues, (b) EDX spectrum of the area within the white circle; (c-d) SEM images at different magnifications (see the footers for experimental settings).	5
Figure S5. (a) SEM images at different magnifications and EDX spectrum of 1-ED@ITO as-prepared and cleansed with MilliQ water to remove salt residues, (b) SEM images at different magnifications and EDX spectrum of 2-ED@ITO as-prepared (see the footers for experimental settings). Insets on the top: CA currents at +1.5 V <i>vs.</i> AgCl/Ag by using these electrodes.	6
Figure S6. (a) SEM images and EDX spectrum of the as-prepared 1-DIP-Nf@ITO sample and (b) the same sample after the electrochemical investigations (for details of the follow-up electrochemistry see Fig. 4a-c). The SEM parameters are found in the footers above.	7

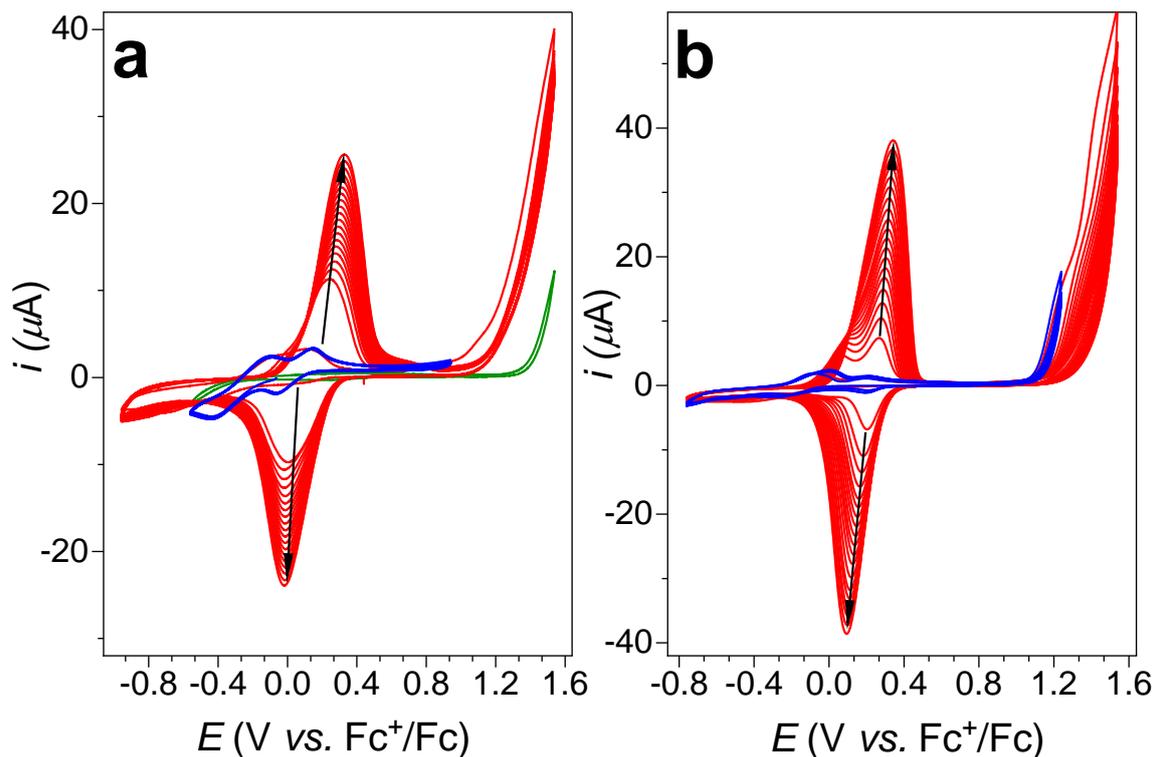


Figure S1. (a) CV scans of **1** (0.4 mM) in DCM with added H₂O (3 mmol in 0.5 mL acetone) on a BDD electrode (0.071 cm²); (b) CV scans of **2** (0.4 mM) under the same conditions. Settings: under Ar, $\nu = 100$ mV/s, Pt, and Ag⁺/Ag (0.01 M AgNO₃, 0.1 M TBAP/acetonitrile), as counter, and reference electrodes, respectively. Red curves: 20 cycles in a broader potential range in both cases show that the anodic oxidation above +1.2 V *vs.* Fc⁺/Fc is required for electrodeposition (compare to the blue CVs, 10 cycles, in a narrower potential range). The green CV was recorded in DCM (10 mL) with added H₂O (3 mmol in 0.5 mL acetone) on a BDD electrode (0.071 cm²) for comparison.

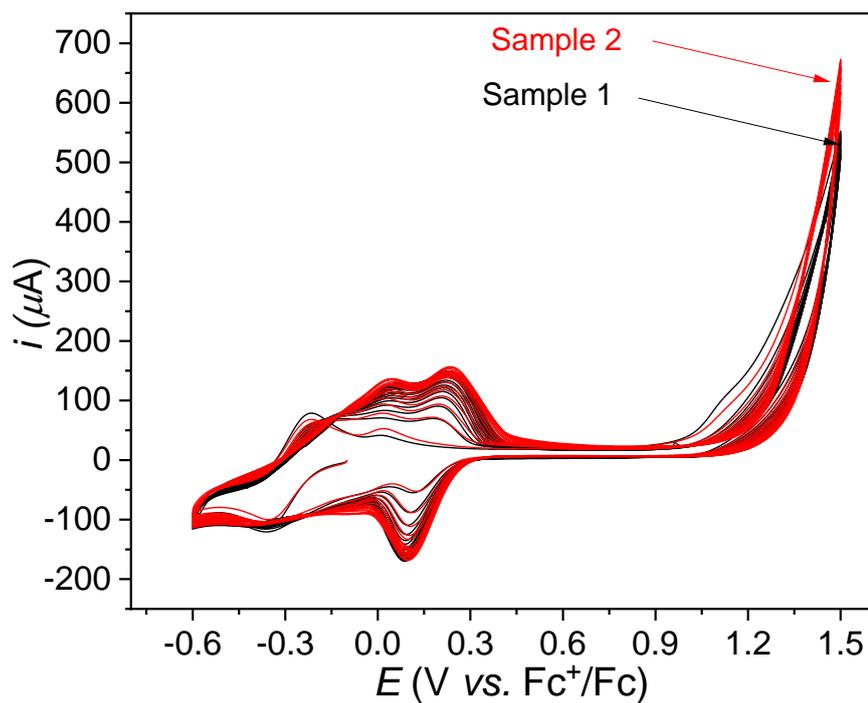


Figure S2. Comparison of two parallel electrodeposition experiments using complex **1** (0.8 mM) on ITO (1.5 cm²) in DCM with added water (3 mmol in 0.5 mL acetone) illustrating good reproducibility. Settings: under Ar, $\nu=100$ mV/s, 20 cycles, Pt, and Ag⁺/Ag (0.01 M AgNO₃, 0.1 M TBAP/acetonitrile), as counter, and reference electrodes, respectively. See the analogous experiments on FTO in Fig. 3c for comparison.

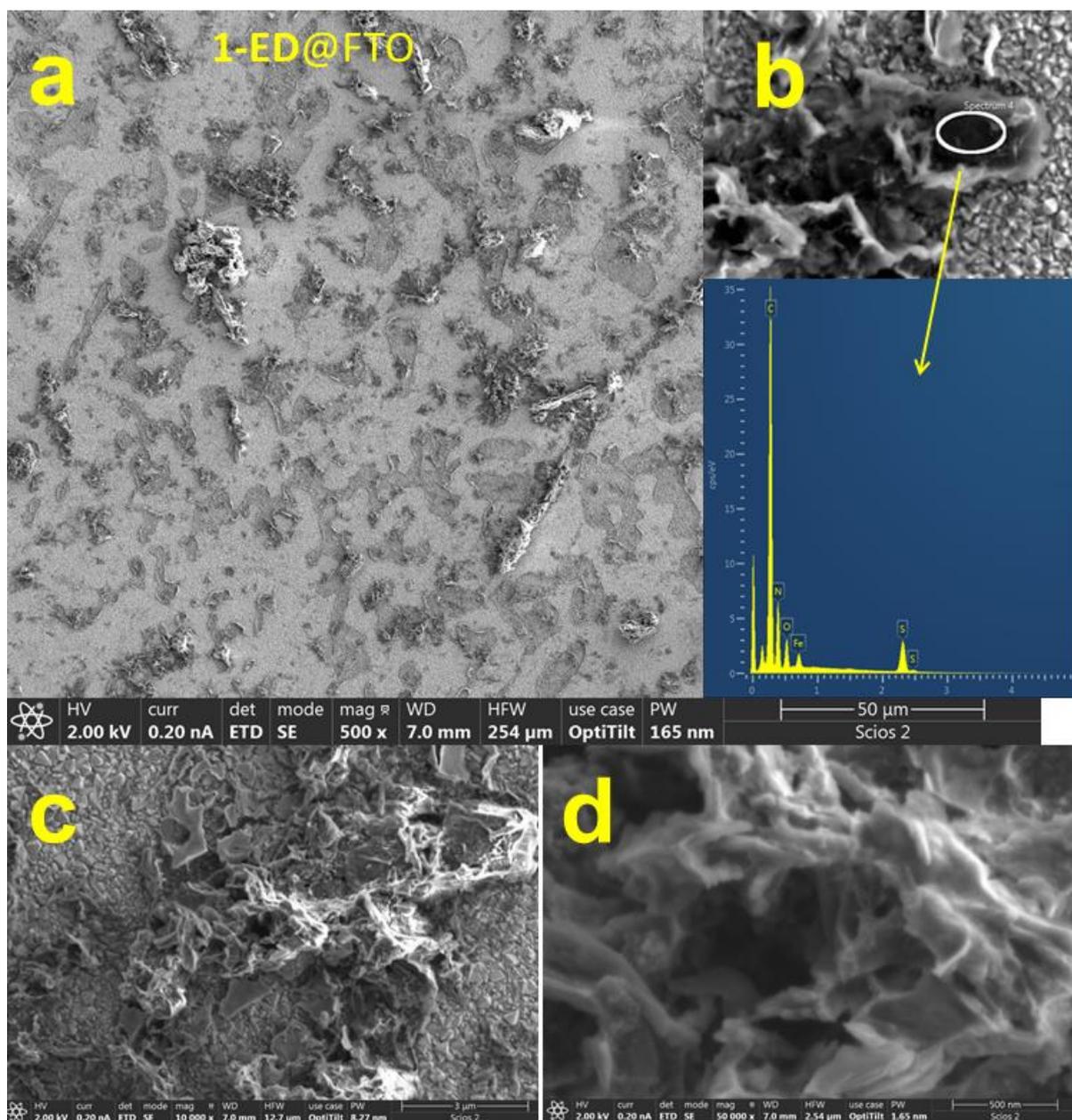


Figure S3. (a) SEM image of 1-ED@FTO as-prepared and cleansed with MilliQ water to remove salt residues, (b) EDX spectrum of the area within the white circle; (c-d) SEM images at different magnifications (see the footers for experimental settings).

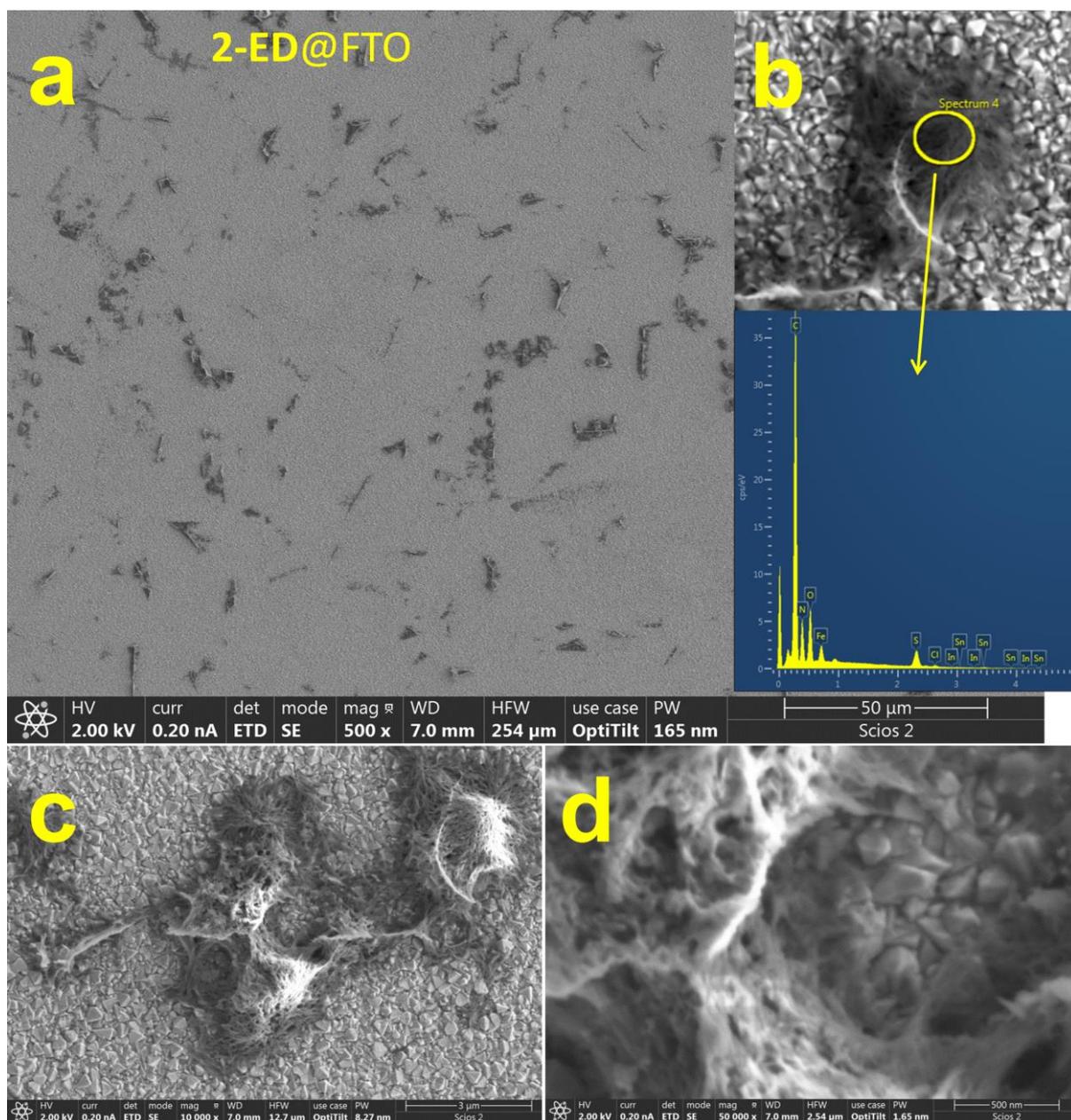


Figure S4. (a) SEM image of 2-ED@FTO as-prepared and cleansed with MilliQ water to remove salt residues, (b) EDX spectrum of the area within the white circle; (c-d) SEM images at different magnifications (see the footers for experimental settings).

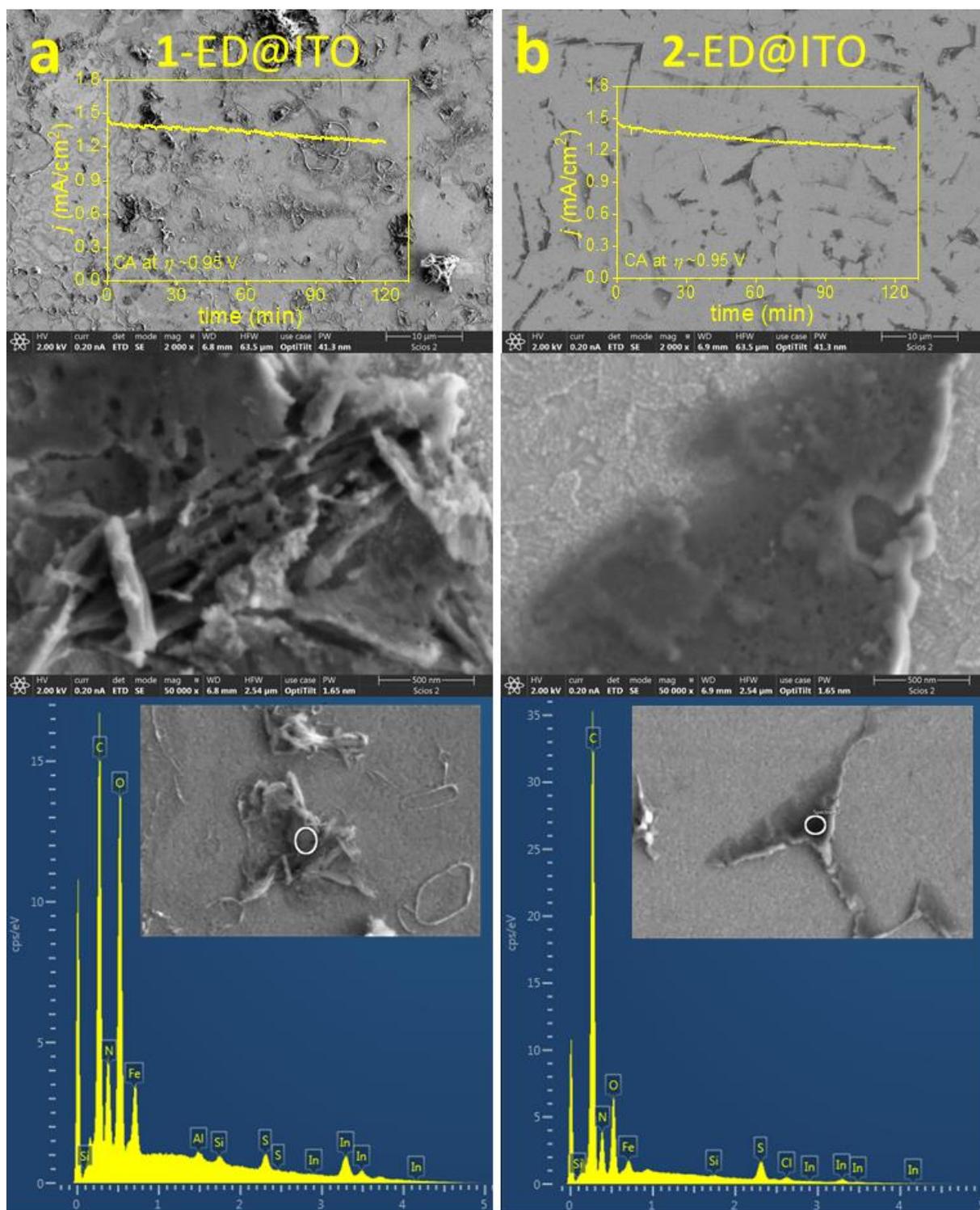


Figure S5. (a) SEM images at different magnifications and EDX spectrum of 1-ED@ITO as-prepared and cleansed with MilliQ water to remove salt residues, (b) SEM images at different magnifications and EDX spectrum of 2-ED@ITO as-prepared (see the footers for experimental settings). Insets on the top: CA currents at +1.5 V *vs.* AgCl/Ag by using these electrodes.

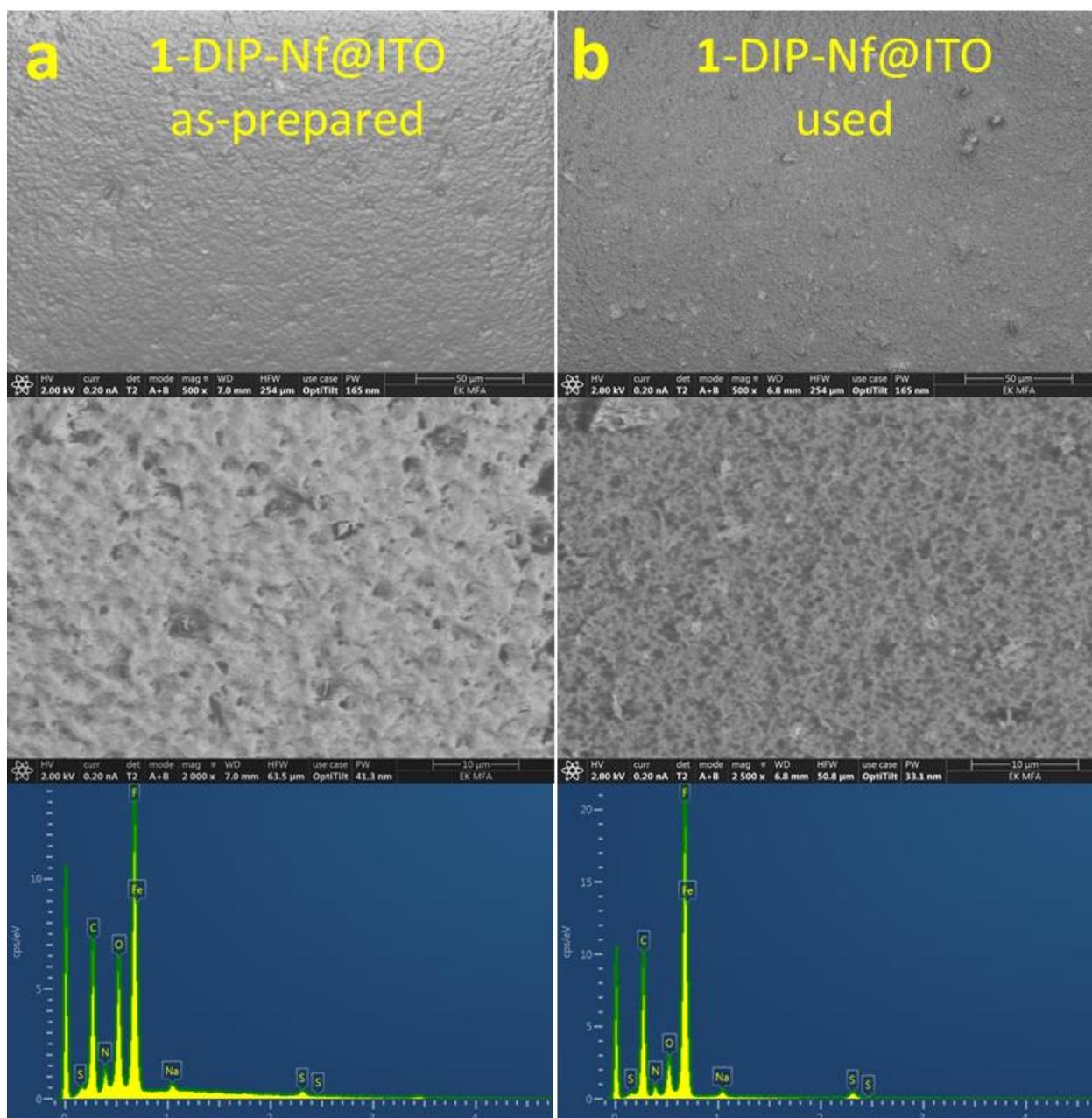


Figure S6. (a) SEM images and EDX spectrum of the as-prepared 1-DIP-Nf@ITO sample and (b) the same sample after the electrochemical investigations (for details of the follow-up electrochemistry see Fig. 4a-c). The SEM parameters are found in the footers above.