Supporting information

Charge-storage mechanism of highly defective NiO nanostructures onto carbon nanofibers in electrochemical supercapacitor

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Raman shift (cm⁻¹)

Figure S1. (a) Topview SEM micrograph and (b) Raman spectrum of the as-prepared CNFs.



Figure S2. Schematic representation of the electrochemical cell used in the *operando* Raman studies.



Figure S3. XRD pattern of (a) CNFs and (b) NiO@CNFs: (*i*) as-prepared materials and (*ii*) wetted electrodes in a $1 \text{ m Li}_2\text{SO}_4$ aqueous electrolyte. P.S. Wetted XRD patterns were taken with synchrotron light in order to increase the photon flux.



Figure S4. Cyclic voltammograms of the NiO electrode housed in a conventional threeelectrode cell. Electrolyte: $1 \text{ m Li}_2\text{SO}_4$ aqueous solution. Scan rate (a) 1 mV s^{-1} and (b) 1 to 100 mV s^{-1} .



Figure S5. Contrasting GCD from (i) CFNs and (ii) NiO@CNFs at 5 mA.



Figure S6. Raman spectra of NiO@CNFs: (*i*) as-prepared and (*ii*) wetted materials in a $1 \text{ M Li}_2\text{SO}_4$ aqueous electrolyte: (**a**) zooming in the 300 to 800 cm⁻¹ range and (**b**) full range spectra for overview. No voltage was applied to cell here (unplugged cell, OCV).



Figure S7. Extended Raman spectra of the CNFs electrode under dynamic polarization conditions in a $1 \text{ M Li}_2\text{SO}_4$ aqueous electrolyte from 0 to 1.0 V and with 633 nm excitation laser wavelength.



- Li₂SO₄.H₂O

Figure S8. Raman spectra of CNFs electrode under polarization conditions in a 1 M Li₂SO₄ aqueous electrolyte from 1.0 a 0 V and with 633 nm excitation laser wavelength zooming at 300 to 800 cm⁻¹ range.



Figure S9: Raman spectra of the CNFs electrode under polarization conditions in 1 M Li₂SO₄ aqueous electrolyte from 1.0 to 0 V and with 633 nm excitation laser wavelength zooming at 950 to 1200 cm⁻¹ range.