Electronic Supplementary Information

Sustainable one-pot aqueous route to hierarchical carbon-MoO₂ electrodes for Li-ion batteries

Julie Besnardiere^{a,b,c}, Xavier Petrissans^{a,b}, Christine Surcin^e, Valérie Buissette^d, Thierry Le Mercier^d, Mathieu Morcrette^e, David Portehault^{a,b,c,*} and Sophie Cassaignon^{a,b,c}

Email: david.portehault@upmc.fr

^a Sorbonne Universités, UPMC Univ Paris 06, UMR 7574, Chimie de la Matière Condensée de Paris, F-75005, Paris, France.

^b CNRS, UMR 7574, Chimie de la Matière Condensée de Paris, F-75005, Paris, France.

^c Collège de France, Chimie de la Matière Condensée de Paris, 11 place Marcelin Berthelot, 75231 Paris Cedex 05, France.

^d Solvay Research & Innovation, Paris Center, 52 rue de la Haie-Coq, 93308 Aubervilliers Cedex, France.

^e Laboratoire de Réactivité des Solides, UMR CNRS 7314, Université de Picardie Jules Verne, 33 Rue Saint Leu, 80039 Amiens Cedex, France.



Figure S1 FESEM image of (a) as-synthesized organic cores as a reference made by hydrothermal treatment of an ascorbic acid solution (0.15 mol.L⁻¹, pH 1, 12 h at 250 °C). Even at low tension (1 kV), charge effects are visible, due to the low conductivity of the organic core before calcination; (b) after calcination (7h, 600 °C under argon). The apparatus tension was lowered from 5 kV to 1 kV to register picture (a) before calcination because of charge accumulation in the non-conductive sample.



Figure S2 UV-visible-near IR absorbance spectra of the C-MoO₂ particles (black) compared to bare 30 nm MoO₂ nanoparticles (grey). The noise at *ca*. 900 nm corresponds to a change in the UV-vis. lamp. The absorbance was calculated from the original reflectance and wavelength coordinates of the spectra and transformed to Kubelka-Munk function F(R) versus photon energy, where F(R) is considered as the absorbance.



Figure S3 XRD monitoring of MoO_2 crystallization during the carbon- MoO_2 core-shell formation. The reaction is stopped after 1 h (a1 and a2), 3 h (b), 6 or 12 h (c) of hydrothermal treatment. The difference between the two 1h-samples is due to the fact that for such short times, it is difficult to master the hydrothermal treatment duration. 6 h and 12 h samples present the same XRD profile. a star (*) indicates the peaks belonging to monoclinic MoO_2 according to the reference pattern (ICDD 04-003-1961. The most intense peaks are indexed.



Figure S4 Fourier-Transform Infrared spectra recorded in ATR (Attenuated Total Reflectance) mode, for different materials: (a) the reference core (ascorbic acid at pH 1, hydrothermally treated at 250°C for 12 hours). (b) Reference MoO₂ nanoparticles without the organic core, made by using carbon-free hydrazine reductant. (c) The as-synthesized carbon-MoO₂ core-shell product. (d) The calcined carbon-MoO₂ core-shell product.



Figure S5 FESEM images of the C-MoO₂ sample (a) before and (b) after SPS (Spark Plasma Sintering) treatment.



Figure S6 (a) XRD patterns of the core-shell assemblies before (dotted line) and after (solid line) calcination. The patterns can be fully indexed to the MoO₂ monoclinic structure. (b) and (c) FESEM images of the core shell structures before and after calcination, respectively.