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Comodification of commercial TiO2 anode by combining solid electrolyte with pitch-derived carbon to boost cyclability and rate capabilities

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1. Characterization

Crystal phases were analyzed by a Rigaku Dmax-2500 diffractometer with Cu Ka radiation as the excitation source. Carbon content was studied by Thermogravimetric analysis (TGA) by heating at 10 ºC $min^{-1}$from room temperature to 800 ºC in air. High-resolution transmission electron microscope (HRTEM, JEOL JEM-2100) was used to scrutinize microstructures. High angle annular dark field (HAADF) scanning transmission electron microscope (STEM) examination and energy-dispersive X-ray spectroscopy (EDS) experiments were carried out on a 100 kV microscope (JEOL JEM-2200FS) equipped with a Bruker Quantax 400 EDS system with an XFlash 5060 detector. Graphitization degree of carbon was characterized by a Renishaw cofocal Raman microspectroscopy (Renishaw Co. Ltd., Gloucestershire, U.K.) system with a laser excitation wavelength of 780 nm. X-ray

photoelectron spectra (XPS) were recorded on a KARTOS XSAM800 X-ray photoelectron spectrometer (Kratos Analytical Ltd., Manchester, U.K.) using Al Kα radiation (hν = 1486.6 eV) as the excitation source (V = 12 kV, I = 10 mA).

2. Electrochemical Test

The assembly of coin cells was conducted in a glovebox full of Ar gas atmosphere. To obtain the working electrodes, the homogeneous slurry comprised of 80 wt % active materials, 10 wt % carbon black, and 10 wt % binder containing 9.0 wt % polyvinylidene difluoride solved in n-methyl pyrrolidinone were coated on copper foil current collectors and subsequently dried for 12 h at 120 °C under vacuum. The copper foil loaded with the slurry was punched into the electrode slices (14 mm in diameter with active material loading around 3 mg). In addition, Li foil was used as counter electrode in a CR2025 coin cell, and Celgard 2300 was employed as separator. Electrolyte is a mixture of ethylene carbonate/dimethyl carbonate (1:1 by volume) dissolving 1 M LiPF6. The galvanostatic discharge−charge were tested by the battery test system of Land CT2001A at room temperature from 0.02 to 3.0 V (vs Li/Li^+). An IviumStat electrochemistry workstation was adopted to carry out cyclic voltammetry (CV) tests at charge/discharge rate of 0.2 mV s^{-1}. Electrochemical impedance spectroscopy (EIS, with an AC signal amplitude of 5 mV between 100 kHz and 0.01 Hz) measurement was performed on the same working station to the CV test.

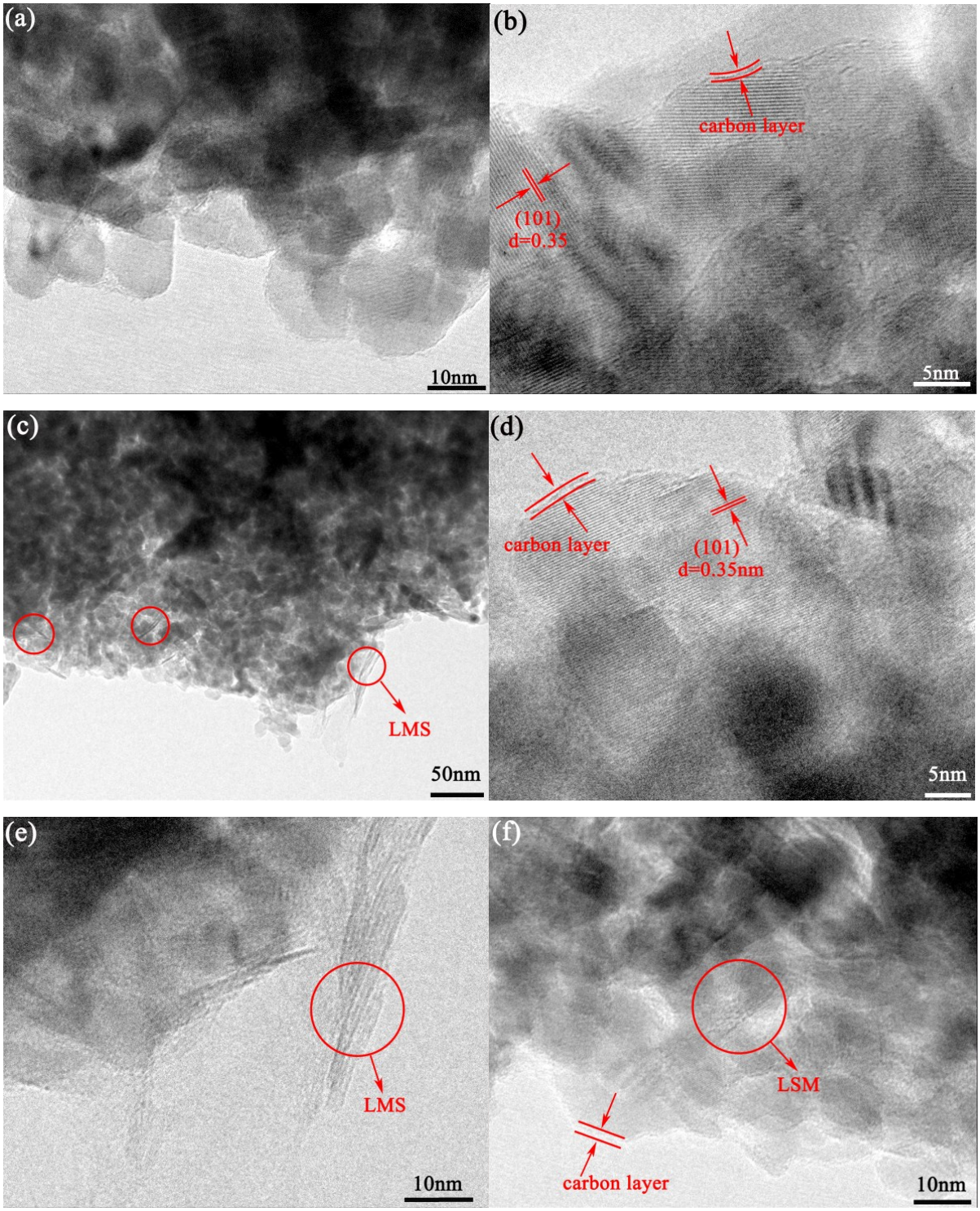

Fig. S1 TEM images of LSM1 (a, b) and LSM3 (c-f).

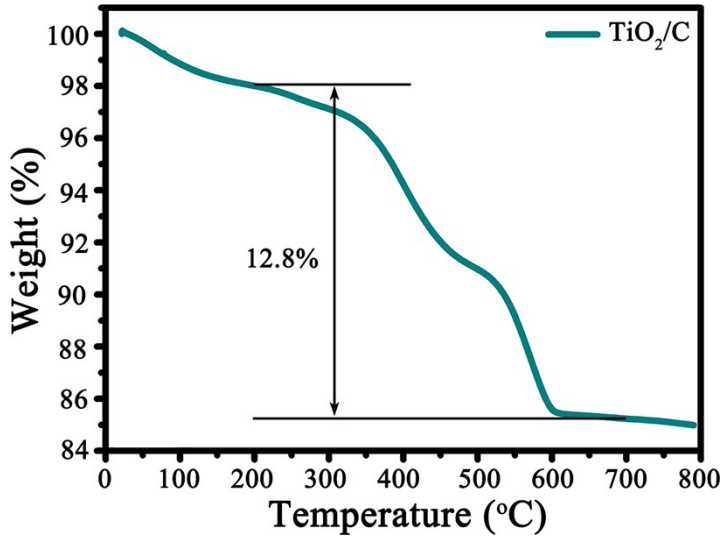

Fig. S2 TG curves of TiO_2/C.

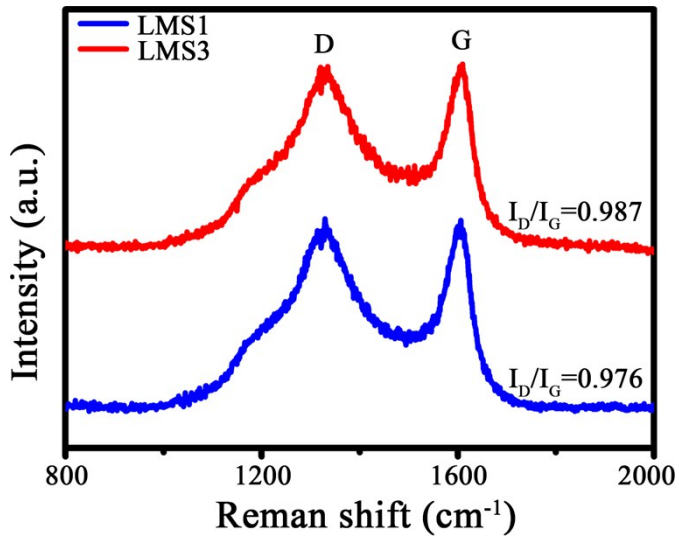

Fig. S3 RS of LMS1 and LMS3.

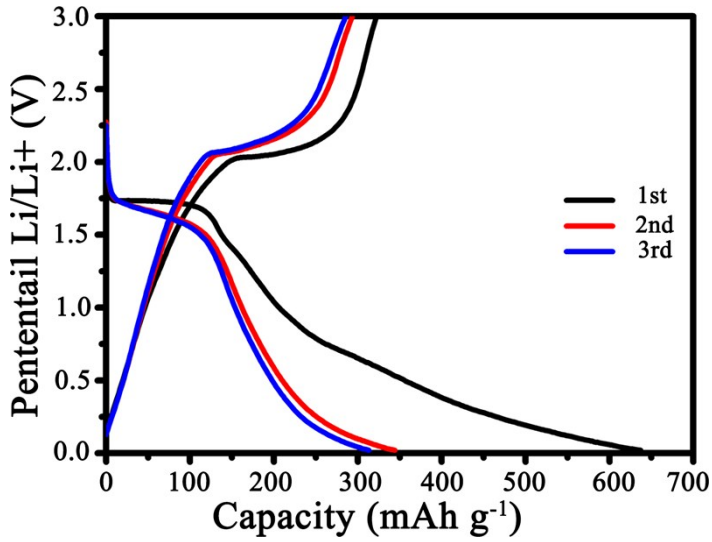

Fig. S4 Lithiation/delithiation curves at 100 mA g^{-1} for LMS2.

Table S1 Comparison of the electrochemical performance between the comodified c-TiO_2 in this work and carbon-coated TiO_2 anodes in the same potential range of 0.01-3V

Active material	Current density – capacity – cycle number – capacity retention (relative to the 2nd cycle)
TiO_2@C/RGO [S1]	800 mA g^{-1} – 158.6 mAh g^{-1} – 500 – >100%
TiO_2/C [S2]	500 mA g^{-1} – 140.2 mAh g^{-1} – 1000 – <70%
TiO_2/RGO [S3]	420 mA g^{-1} – 200 mAh g^{-1} – 500 – >100%
C-TiO_2 [S4]	335 mA g^{-1} – 159.8 mAh g^{-1} – 960 – ~90%
TiO_2(B)@carbon [S5]	750 mA g^{-1} – 200 mAh g^{-1} – 5 – >100%
TiO_2/N-doped carbon [S6]	305 mA g^{-1} – 290 mAh g^{-1} – 100 – ~50%
TiO_2/CS7 [S7]	500 mA g^{-1} – 149.1 mAh g^{-1} – 1000 – 89%
TiO_2/GO [S8]	500 mA g^{-1} – 165 mAh g^{-1} – 20 – >90%
TiO_2@C [S9]	500 mA g^{-1} – 155.8 mAh g^{-1} – 800 – >100%
TiO_2/C [S10]	100 mA g^{-1} – 225 mAh g^{-1} – 100 – <60%
LMS2 (this work)	500 mA g^{-1} – 245 mAh g^{-1} – 1000 – >100%

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