Supporting Information

Synthesis of functionalized 3D hierarchical porous carbon for high-performance supercapacitor

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Figure S1. SEM images of THPC.



Figure S2. SEM images of AC (obtained through annealing the mixture of PPy and KOH at 700 °C).



Figure S3. Raman spectrum of PPy microsheets.



Figure S4. Nitrogen adsorption-desorption isotherms and pores size distribution (PSD, inset) of PPy microsheets.

As shown in **Fig. S3**, PPy microsheets show a type-II adsorption-desorption isothermal, which is typical for non-porous or macroporous adsorbent.¹ The pores size distribution (PSD, the inset of **Fig. S3**, calculated via BJH method by using nitrogen adsorption data) of PPy reveals the existence of a small amount of micropores and macropores, which arise from the interstitial pore space in particle aggregates. Calculated by the Brunauer-Emmett-Teller (BET) model, the specific surface area of PPy microsheets is 7.37 m² g⁻¹.



Figure S5. Survey XPS spectra of THPC.

As obtained using an element analyzer, THPC is composed of 77.0 wt.% C, 12.4 wt.% O, 7.7 wt.% N, 1.2 wt.% S and 1.5 wt.% H.



Figure S6 Electrochemical performances of THPC as anode electrode for lithium ion batteries: (a) discharge/charge curves at 0.2C; (b) capacity over cycling at different rates; (c) cyclability and coulombic efficiency at 5C; (d) impedance spectra of THPC after the 1st, 10th, 50th, 100th and 500th cycle at 5C.

THPC was also tested as anode electrode for lithium-ion batteries. Benefiting from its unique 3D hierarchical porous nanostructure and high-level N and O-doping, the THPC exhibits superior rate and cycling performances. As shown in **Figure S6b**, the reversible capacities are 1269, 944, 671, 553, 341 and 259 mAh g⁻¹ at 0.2, 1, 5, 10, 30 and 50 C (1 C = 372 mAh g⁻¹), respectively. When cycling at 5 C, the capacity is initially 344 mAh g⁻¹, gradually increases to 921 mAh g⁻¹ after 600 cycles and maintains up to 907 mAh g⁻¹ even after 1000 cycles (**Figure S6c**). The increasing capacity is due to the improvement of surface wetting between the electrode and electrolyte during extended cycling and the electroactivation process (**Figure S6d**). The obtained Li⁺-ion storage capability is extremely high among the carbonaceous anode materials.²⁻⁴

References

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