Supporting Information

Constructing graphitization graded porous carbon using olive leaves as a carbon source for high-performance zinc-ion hybrid capacitor

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Experiment Section

Fabrication of OLPCs from wasted olive leaves: The olive leaves were washed several times with distilled water (DL) to remove the surface dust, dried under blast at 100 °C for 12 h The leaves were then crushed and ground to powder. Then it was pre-carbonized in nitrogen atmosphere at 600 °C for 2 h to obtain olive leaf carbonate (OLPC). OLPC was mixed with activator at different mass ratios (KOH: C = 1:1, 2:1, 3:1). A little DL was added and ultrasonicated to make the mixture completely homogeneous, then most of the water was evaporated by magnetic stirring at constant temperature. After drying, it was transferred to a tube furnace and annealed at 750 ℃ for 2 h in a nitrogen atmosphere (heating rate: 5 °C/min). The annealed product was etched with 2 M HCl, then washed sufficiently with DL to neutral, and dried at 100 °C for 12 h to obtain OLPC. The product was named OLPC-X, and X was the mass ratio of KOH to carbon. That is, OLPC-1, OLPC-2, OLPC-3. The initial mass of olive leaves is 0.248 g, and the OLPC obtained after pyrolysis at 750 ℃ is 0.213 g. The production yield of carbon materials is 85.9 %.

Materials characterization

The crystal structures of the samples were studied by X-ray diffrac-tometer (XRD, Shimadzu-7000, λ=0.1541 nm, 40 kV). In addition, the elemental composition of the sample was characterized by X-ray photoelectron spectroscopy (XPS, Thermo Scientific K-Alpha). N2 ab-sorption and desorption isotherms (JW-TB200) were used to study their specific surface area and pore size distribution by the Brunauer-Emmett-Teller (BET) method. The morphology and microstructure of the samples were

observed by scanning electron microscope (SEM, Gemini ZEISS Sigma 300) and high-resolution transmission electron microscope (JEM-2100 plus). The wettability of the electrodes was measured using a contact angle tester (SDC-350KS).

Electrochemical characterization

The active material (OLPC), acetylene black and polyvinylidene fluoride (PVDF) were mixed homogeneously into N-methyl-2-pyrrolidone (NMP) at a mass ratio of 8:1:1. The above paste was pressed onto a carbon paper with a diameter of 1 cm and dried under vacuum at 60 °C for 12 h. Then, the CR2032 button batteries are assembled using zinc foil as anode and the as-obtained sample as cathode. The electrolyte was weakly acidic 2 M $ZnSO_4$. The average loading mass of cathode is 1.3-1.8 mg cm^{-2}. An automated battery tester (Neweare, CT-4008 T) was used to test the electrochemical performance of the batteries. Cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) tests were also performed on a CHI660E electrochemical workstation. The wettability of the electrodes was measured using a contact angle tester (SZ-CAMC32).

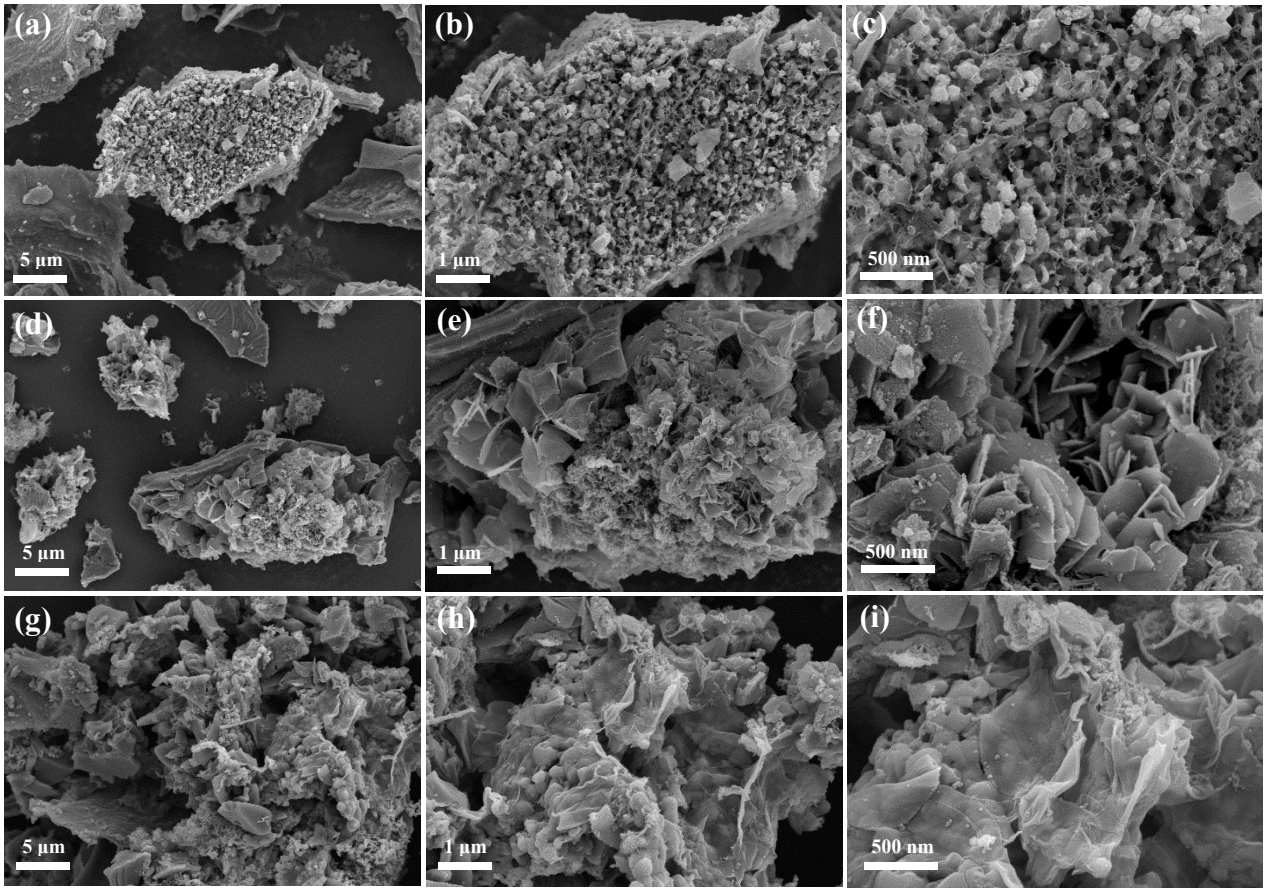

Figure S1 SEM images of (a-c) blank material, (d-f) OLPC-1 and (g-i) OLPC-3

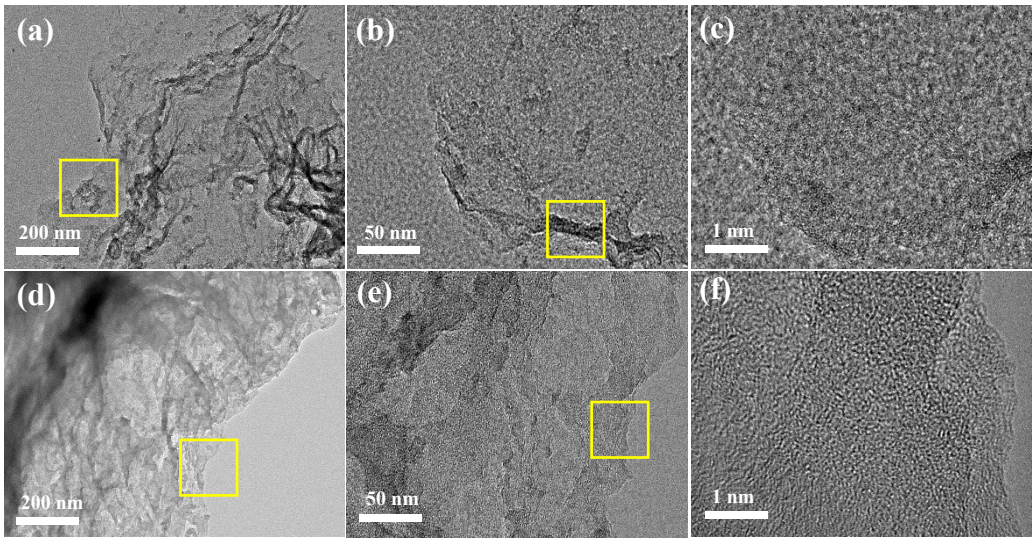

Figure S2 TEM images (a-c) OLPC-1 and (d-f) OLPC-3

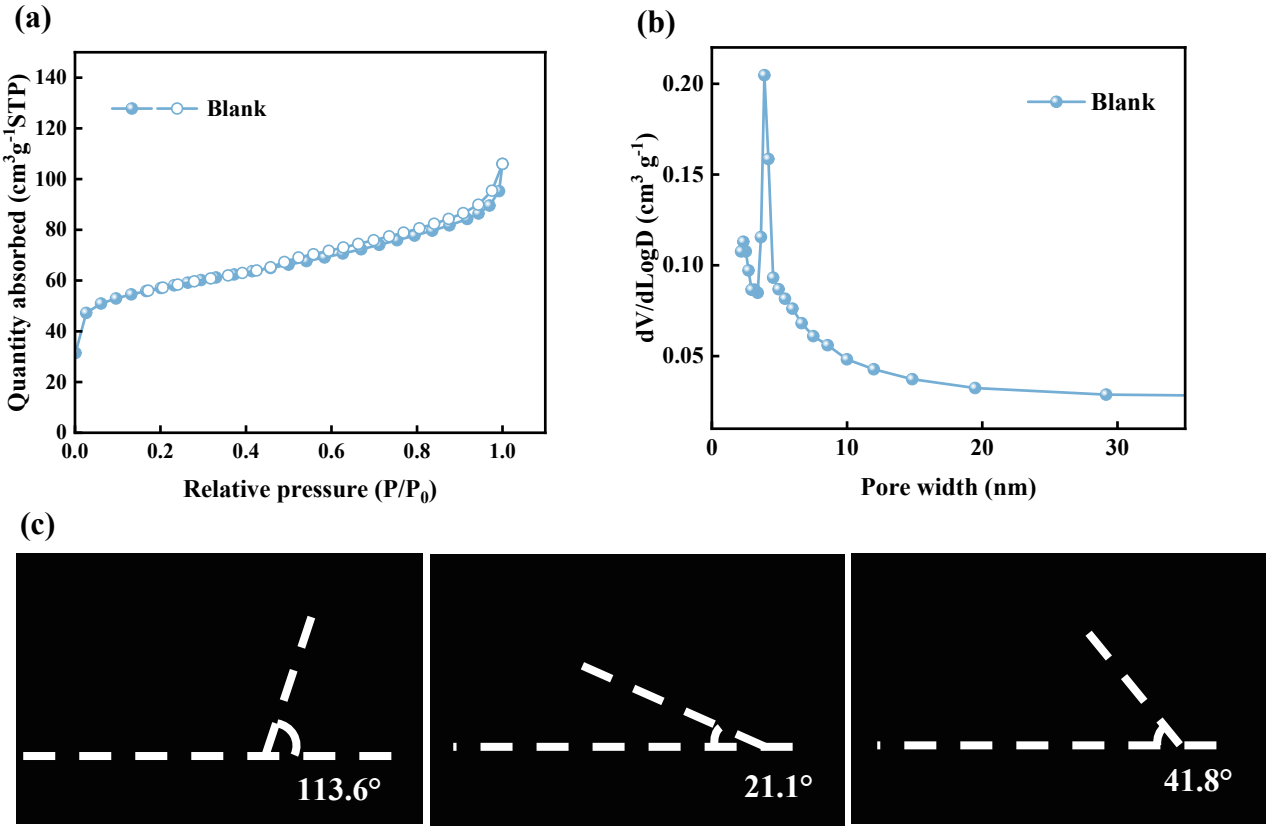

Figure S3 (a) N_2 adsorption/desorption isotherm (b) Pore size distribution curve

(c) Water contact angles of wasted OLPC materials

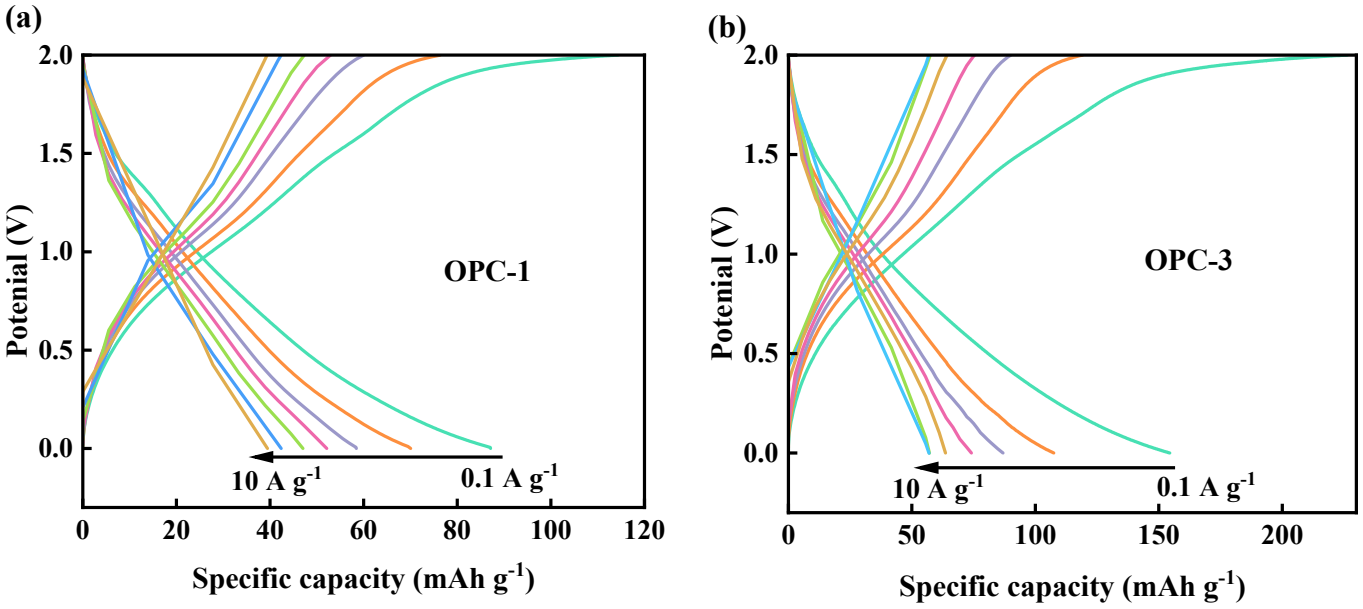

Figure S4 GCD curves (a) OLPC-1 sample (b) OLPC-3 sample

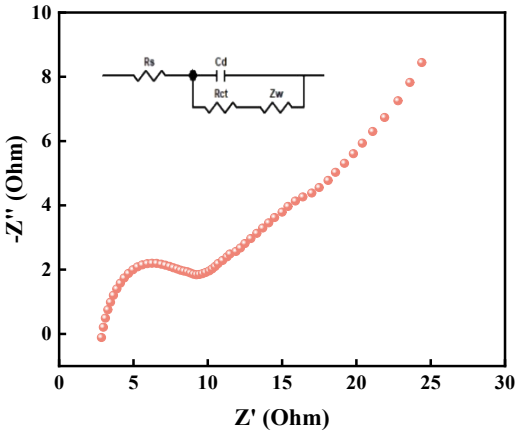

Figure S5 EIS spectra of the OLPC-2-built quasi-solid ZHSC device

Table S1 Specific surface area, total pore volume and mean pore diameter of synthetic samples with different activation temperatures

Samples	Specific Surface Area [m^2/g]	Total Pore Volume [cm^3/g]	Average Pore Diameter [nm]
OLPC-2/700℃	1105	0.675	3.258
OLPC-2/750℃	1497	1.017	4.292
OLPC-2/800℃	1286	0.739	3.209

Table S2 Comparison of Zn^{2+} storage capacities of aqueous ZHSCs assembled by different carbon-based cathodes

Materials	Specific capacity (mAh g^{-1})	Current density (A g^{-1})	Energy density (Wh kg^{-1})	Capacity retention, cycles	Refs.
OLCK4	121.7	0.3	94.3	86.7%, 10,000	1
MEHC-3	132.9	0.2	106.4	94.7%, 20,000	2
PHCA	143.7	1.0	129.3	92.0%, 10000	3
$VO_{0.9}$/C	146	1.0	126.5	84.0%, 10,000	4
PCNs-2	149.0	0.2	119.0	91.0%, 10,000	5
OLPC-2	179	0.1	161.1	92.4%, 10000	This work

References

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