Significant enhanced supercapacitor performance of $W_3Nb_{14}O_{44}$ by introducing serine and histidine-functionalized and boron-doped graphene quantum dot

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1. Experimental

1.1. Materials and reagents

Citric acid (CA), serine (Ser), histidine (His) and boric acid (H_3BO_3) were purchased from Sinopharm Chemical Reagent Co., Ltd (Shanghai, China). Ammonium metatungstate and niobium oxalate were purchased from Aladdin. Silver nanowires (Ag NW) in isopropanol (5 mg mL^{-1}) was purchased from Jiangsu XFNANO Materials Tech Co., Ltd. B/GQD was prepared according to reported in the literature. The solid electrolyte (PVA/Li_2SO_4) was prepared by adding 2.0 g polyvinyl alcohol (PVA) and 10 mL of 3 mol L^{-1} Li_2SO_4 solution in 20 mL ultrapure water and then heated at 75°C with stirring until a clear homogeneous solution was formed. Rambutan was purchased from Hainan Province and then rambutan peel was collected, followed by washing in distilled water and freeze-dried for use.

1.2. Material characterization

The morphology and structure of as-synthesized materials was characterized by scanning electron microscope (SEM, JEOL, S-4800) and transmission electron microscope (TEM, JEOL, Jem-2100). The crystal structure and chemical composition of as-synthesized materials was studied by X-ray diffraction (XRD, Bruker D8 ADVANCE, Cu Kα radiation, λ = 0.15406 nm). The valence states of as-synthesized materials were characterized by X-ray photoelectron spectroscopy (XPS, Kratos, Axis supra) with mono chromated Al KR radiation. The band gap of as-synthesized materials was

evaluated by obtaining UV-visible diffuse reflectance spectra by UV-visible spectrophotometer (UV-vis, Shimadzu, UV-3600 Plus). The oxygen vacancies of as-synthesized materials were characterized by the electron paramagnetic resonance spectra (EPR, Bruker EMX PLUS, X-band≈9.8 GHz). The morphology of rambutan peel was characterized by ultra-depth three-dimensional microscope (Keyence, VHX-1000C).

2. Figures and Tables

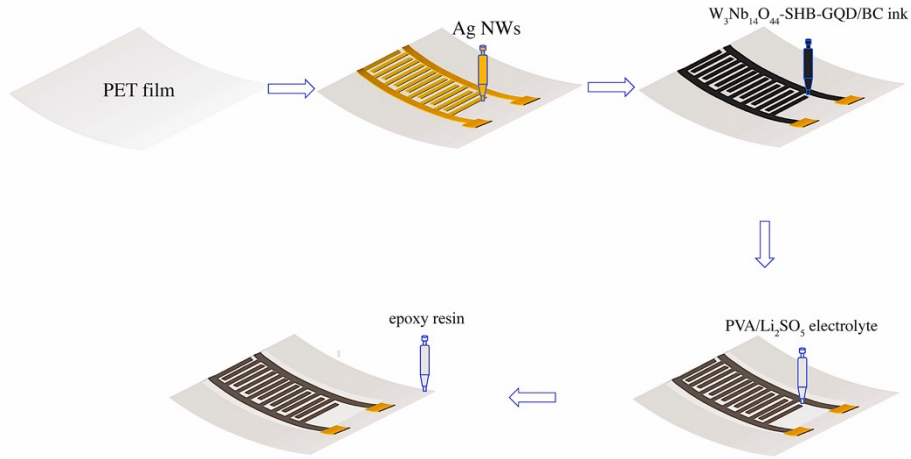

Fig. s1 The procedure for preparation of Flexible symmetrical supercapacitor

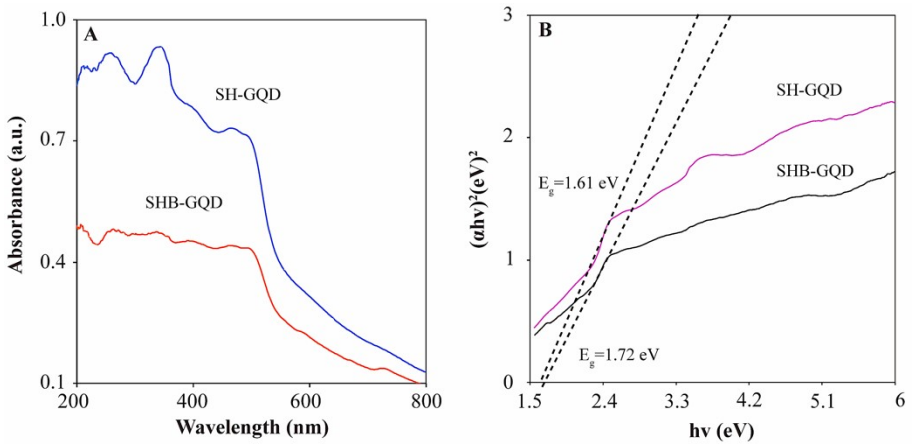

Fig. s2 Diffuse reflectance UV-visible spectroscopies (A) and plots of transformed Kubelka-Munk function *vs*. the energy of light (B) of SH-GQD and SHB-GQD

Fig. s3 Optical photographs of mixed ammonium metatungstate with niobium oxalate (left) and SHB-GQD-W/Nb complex solution (right)

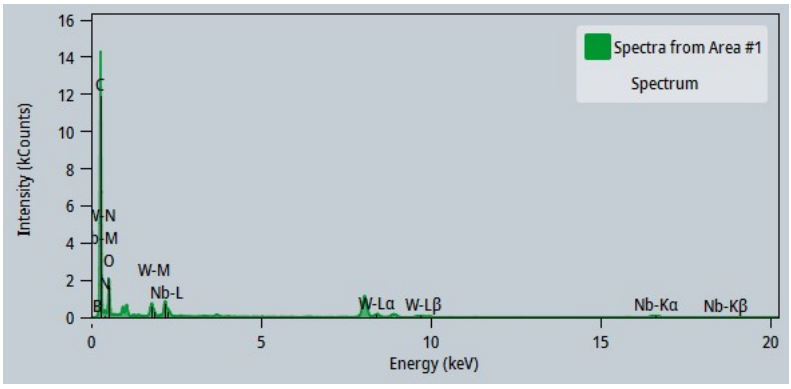

Fig. s4 EDS energy spectrum of $W_3Nb_{14}O_{44}$-SHB-GQD/BC

Table s1 The ratios (I(f)) of XRD peak intensity to the strongest peak intensity for $W_3Nb_{14}O_{44}$.

	2-Theta	h k l	PDF-I(f) ratio	$W_3Nb_{14}O_{44}$-B/GQD-C-I(f) ratio
1	23.629	1 0 1	1	0.828
2	29.169	4 1 1	0.058	0.213
3	32.759	5 2 1	0.676	1
4	34.968	6 1 1	0.153	0.226
5	37.062	3 6 1	0.074	0.818
6	39.059	2 7 1	0.261	0.925
7	42.995	1000	0.045	0.485
8	47.171	8 5 1	0.022	0.393
9	52.800	9 6 1	0.162	0.396
10	58.035	1211	0.222	0.323
11	62.430	1420	0.016	0.213
12	68.392	6131	0.013	0.097

13	69.839	10120	0.002	0.083

Table s2 Element composition in $W_3Nb_{14}O_{44}$-SHB-GQD/BC

Element	Mass Fraction (%)	Atom Fraction (%)
C	70.74	89.11
N	0.33	0.35
O	8.01	7.57
B	0.03	0.042
Nb	14.88	2.42
W	6.01	0.512

Table s3 EIS parameters for different electrodes

Electrode	**$W_3Nb_{14}O_{44}$**	**B/GQD-C**	**$W_3Nb_{14}O_{44}$-B/GQD-C**
R_s (Ω)	9.158	5.499	4.845
C (mF cm^{-2})	0.0000108	0.000252	0.000140
R_{ct} (Ω)	26.76	1.88	0.63
Z_w (Ω)	0.696712	0.075045	0.00042961

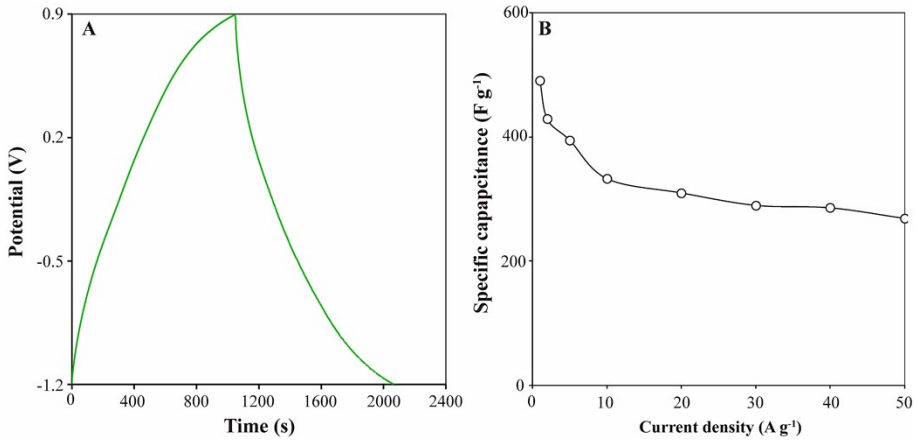

Fig. s5 The charge/discharge curves (E) of $W_3Nb_{14}O_{44}$-SH-GQD/BC electrode at the current density of 1 A g^{-1}, and

specific capacitances (F) of $W_3Nb_{14}O_{44}$-SH-GQD/BC electrode at different current densities