Electronic Supplementary Information

Microfluidic generation of graphene beads for supercapacitor electrode materials

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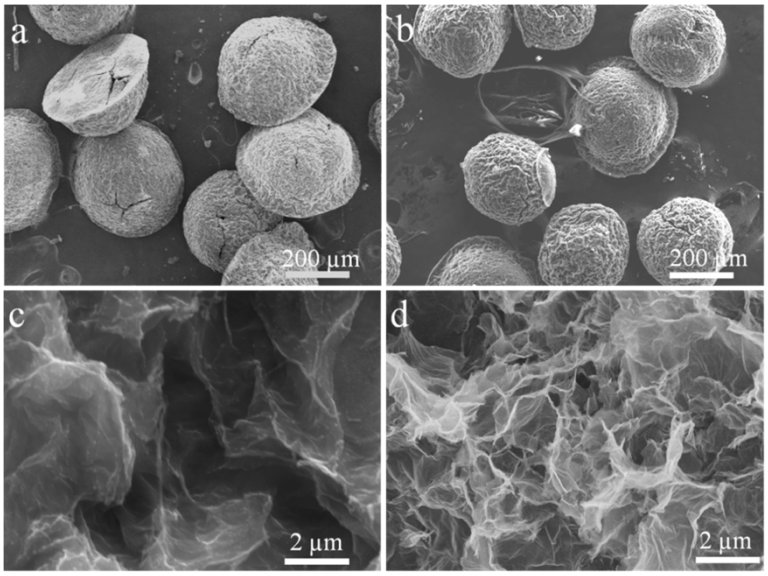

Fig. S1 SEM images of solid GBs prepared by mode a: (a) w (GO) = 0.80%, v_1: v_2 = 1:120, (b) w (GO) = 0.80%, v_1: v_2 = 0.8:120, (c) and (d) were SEM images of their internal and external structure, respectively.

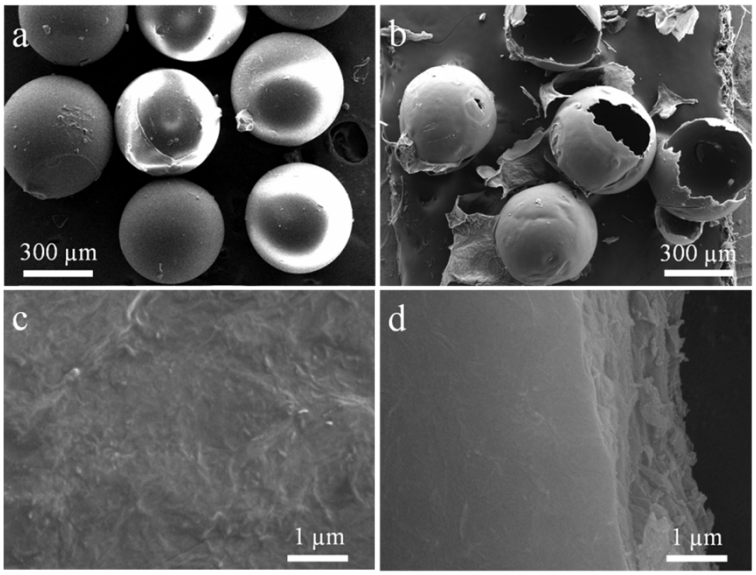

Fig. S2 SEM images of hollow GBs prepared by mode a: w (GO) = 0.1%, v_1: v_2 = 1:120, (a) and (b) were SEM images of hollow GBs, (c) and (d) were SEM images of external and section, respectively.

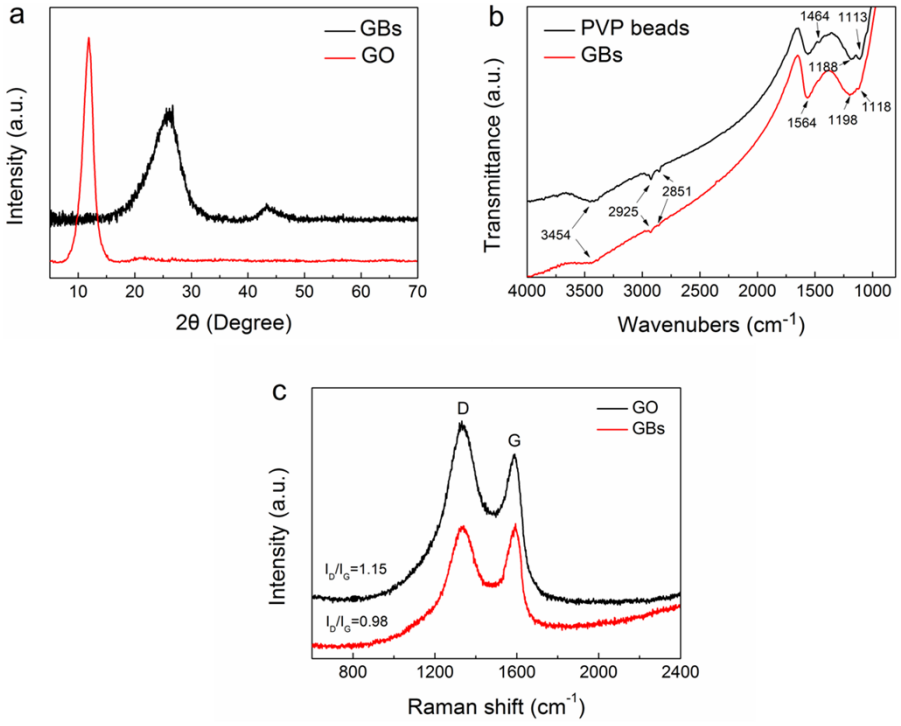

Fig. S3 (a) XRD patterns of GO and GBs. (b) FTIR spectra of PVP beads and GBs. (c) Raman spectra of GO and GBs. GBs were prepared by mode (a), and w (GO) = 0.45%, v_1: v_2 = 0.5:120.

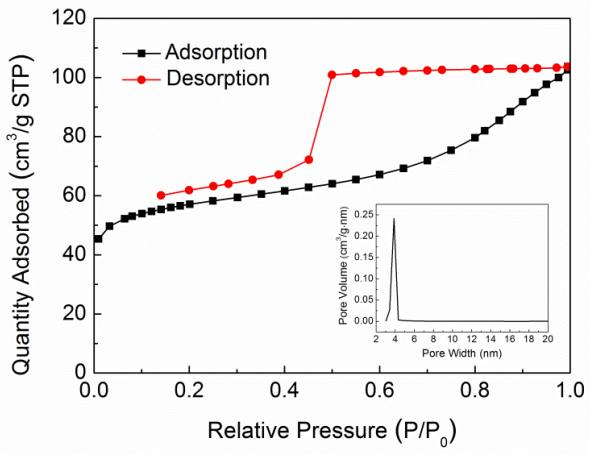

Fig. S4 Isothermal curve for nitrogen adsorption-desorption of the GBs and corresponding pore size distribution (insert picture). GBs were prepared by mode (a), and w (GO) = 0.45%, v_1: v_2 = 0.5:120.

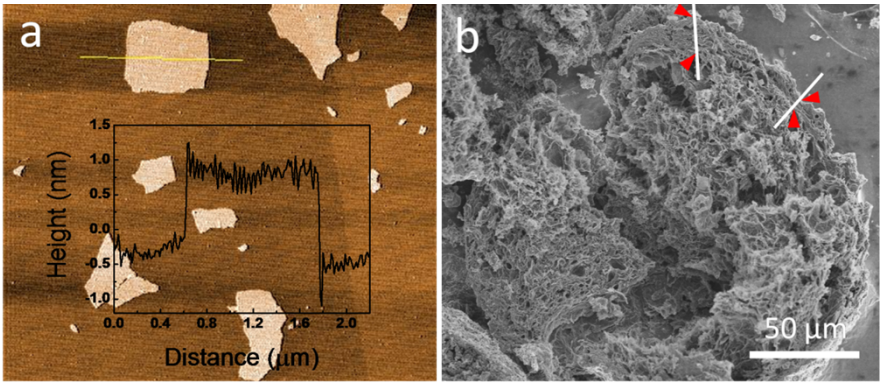

Fig. S5 (a) AFM image (insert was the corresponding height profile) and (b) SEM image of the crushed GBs. GBs were prepared by mode (a), and w (GO) = 0.45%, v_1: v_2 = 0.5:120.

Preparation of PVP beads: PVP solution was consisting of 0.5 g PVP powder and 7.5 mL deionized water. After strong stirring, PVP beads were prepared by the mode (a) of the same device, and pushing speed ratio (v_1: v_2) was 0.5:120.

PVP beads had well spherical structure and uniform size as shown in Fig.S6a. The SEM image of the crushed PVP beads indicated that the beads had hollow structure after the carbonization process.

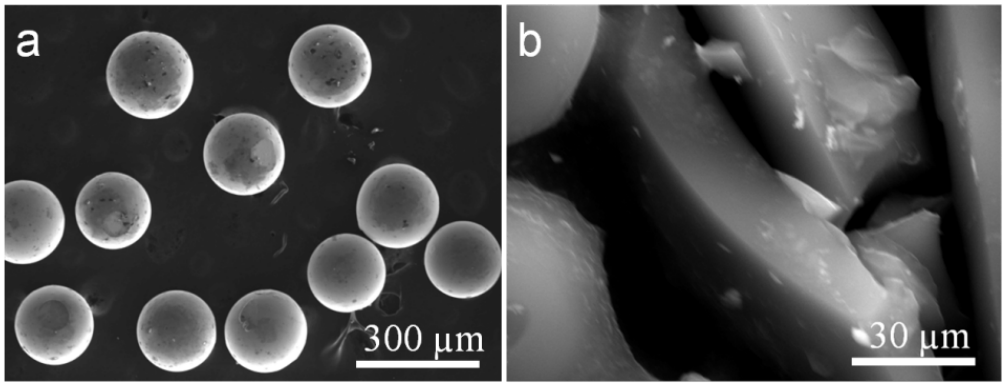

Fig. S6 SEM images of PVP beads (a) and the crushed PVP beads (b).

Fig.S7 was the XRD pattern of PVP beads. It can be seen that there were two peaks in 25.8° and 43°, which corresponded to the graphic (002) and (100) profile, respectively.

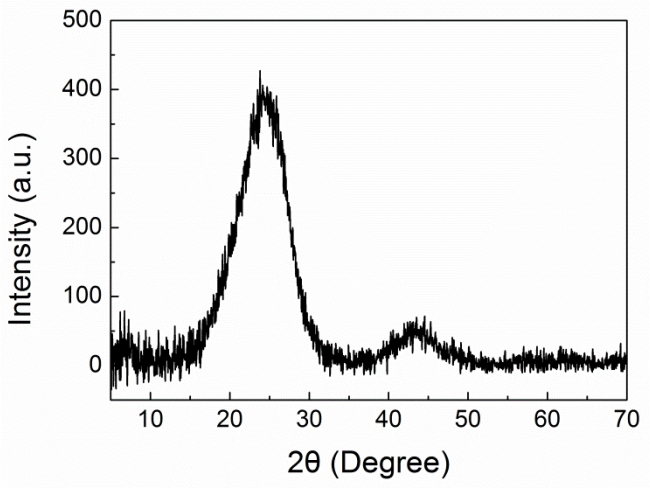

Fig. S7 XRD pattern of PVP beads.

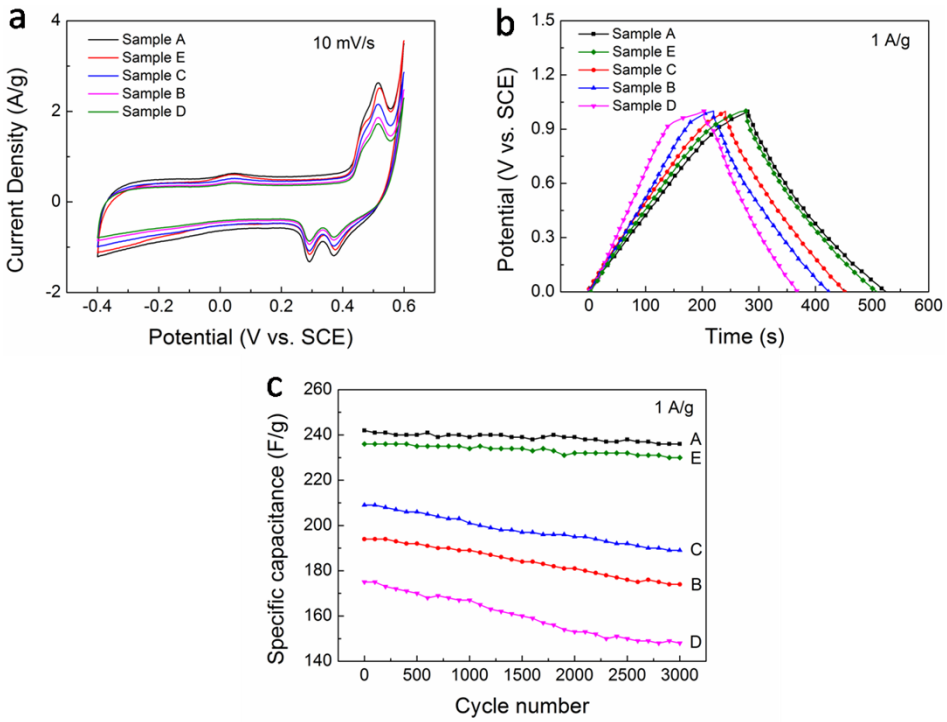

Fig. S8 (a) CV curves of sample A-E at 10 mV/s. (b) GV curves of sample A-E in a current density of 1 $A \cdot g^{-1}$. (c) Charge-discharge cycling test of sample A-E in 1 $A \cdot g^{-1}$. The detail experimental parameters of A-E were shown in Table S1.

Table S1 The electrochemical performance of GBs (sample A-E) with different GO content, the size of droplets and the pushing rate ratio

Sample	solidification method	w (GO)%	v_1:v_2	size (μm)	specific capacitance (F/g)	retention ratio (%)
A	mode a	0.45	0.5:120	184	243	97.5
B	mode a	0.80	1.0:120	335	194	89.8
C	mode a	0.80	0.8:120	298	209	90.3
D	mode a	0.10	1.0:120	387	175	84.7
E	mode b	0.45	0.5:120	162	236	97.9