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In-situ synergistic reduced graphene oxide-boron carbon nitride nanosheets heterostructure for high-performance fabric-based supercapacitors

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Materials and Methods

Materials:

Ultra-pure boron powder (99.99%), urea (99.5%), glucose (99.5%), N, N-dimethylformamide (DMF), polyacrylonitrile (PAN) and poly (vinyl alcohol) (PVA) were obtained from Aladdin Chemical Reagent Co., Ltd. (Shanghai, China). Potassium permanganate ($KMnO_4$), phosphoric acid (H_3PO_4), sulfuric acid (H_2SO_4) and hydrogen peroxide (H_2O_2) were purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). Microfluidic electrospinning device with syringe pumps and Y-snake microfluidic chip was provided by Nanjing Janus New Materials Co., Ltd.

Preparation of rGO-BCN:

0.25 g ultra-pure boron powder, 5 g urea and 0.25 g glucose were uniformly mixed and dried at 80 ℃ for 24 h. The precursors were repeatedly ground for 30 mins to make it uniformly ground. Graphene oxide was prepared by a modified Hummers' method using graphite powder as raw material. 5 g precursor powder was added to 10 mL GO (20 mg mL^{-1}) suspension and dried at 60 °C for 24 h. Then, the dried mixture was transferred to a tube furnace and heated according to the heating procedure set by the tube furnace: heating at 550 ℃ for 3 h under Ar protection flow, then heated at 900 °C for 1 h, and then cooled at room temperature to obtain rGO-BCN (The

yield of BCN is about 9.1%).

Preparation of rGO-BCN fiber membrane electrode:

0.25 g rGO-BCN was dispersed in 5 g DMF by ultrasonic method. 0.6 g of PAN was dissolved in 5 g of DMF and stirred for 3 h. Two solutions of rGO-BCN/DMF (0.7 mL h^{-1}) and PAN/DMF (0.7 mL h^{-1}) were injected into the Y-snake serpentine channel through syringe. A voltage of 18 kV was applied at 2 cm of the needle tip, and the spinning distance of the needle tip was adjusted to 12 cm from the receiver. The spinning was performed at 500 rpm for 5 h to collect the composite nanofiber film. The nanofiber membrane was transferred to a tube furnace and carbonized at 800 ℃ in Ar for 2 h to obtain a flexible rGO-BCN/carbon film electrode.

Preparation of the FSCs:

Preparation of electrolyte H_2SO_4/PVA: 1 g PVA was dissolved in 10 mL H_2O, and heated at 85 ℃ and stirred for 3 h. Then, 1 g H_2SO_4 (98 wt%) was dropped into the above solution and further reacted for 1 h.

Then, the above solution was evenly dropped on the glass slide and dried at room temperature to obtain a H_2SO_4/PVA gel electrolyte membrane which was subsequently placed between the two fiber membrane electrodes and hot pressed at 60 °C for 24 h to obtain FSCs.

Electrochemical measurements:

Cyclic voltammetry (CV), galvanostatic charge-discharge (GCD) and electrochemical impedance spectroscopy (EIS) tests were performed using a CHI760E electrochemical workstation. The calculation formula of mass specific capacitance is: $C_m = (4 \times I \times \Delta t)/(m \times \Delta V)$. Among them, I (A), Δt (s), m (g), ΔV (V) are the discharge current, the discharge time, the mass of the two fiber electrodes and the voltage range, respectively. The calculation formula of power and energy density is as follows: $E = CV^2/8$, $P = E\ \Delta t$, where C, V, Δt are specific mass capacitance, working voltage and discharge time, respectively.

Supplementary Figures

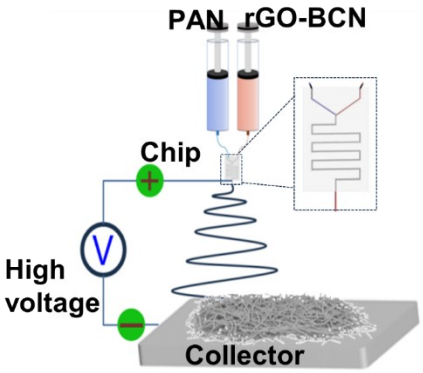

Fig. S1 The preparation of rGO-BCN fiber membrane through MES.

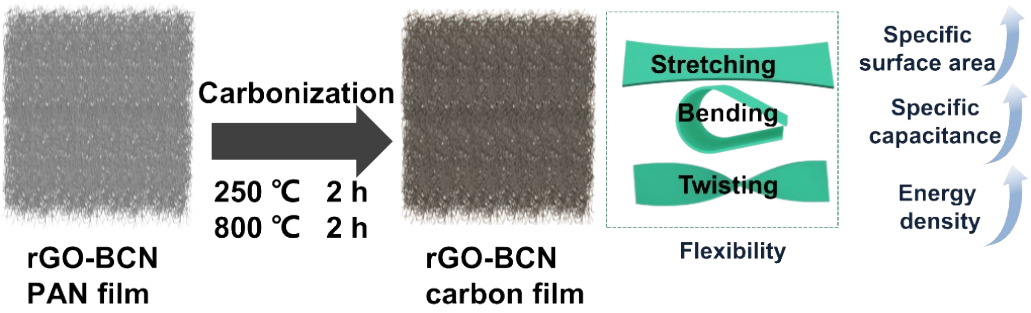

Fig. S2 The carbonization of rGO-BCN fiber membrane.

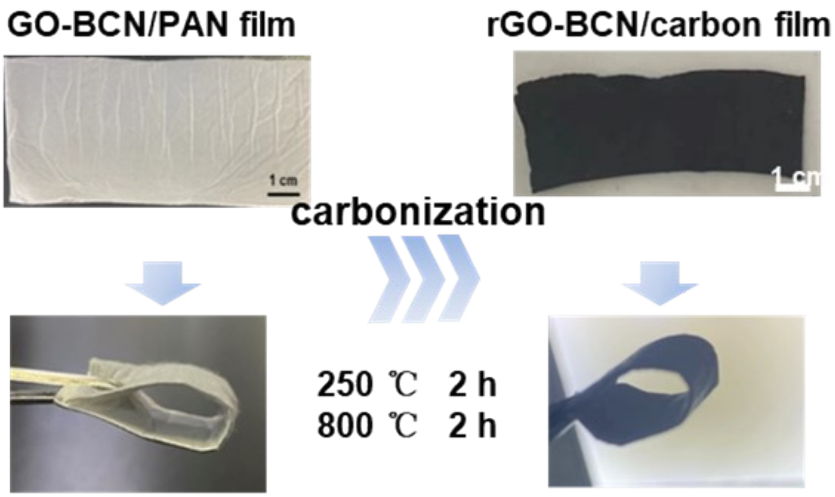

Fig. S3 Photographs of rGO-BCN/PAN film before and after carbonization.

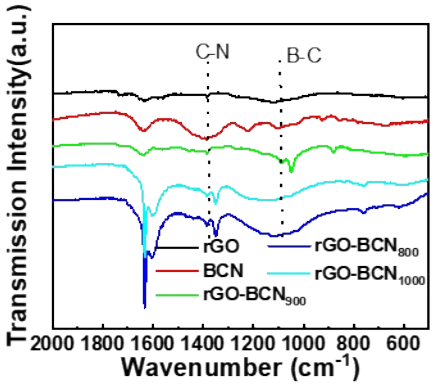

Fig. S4 FTIR spectra of rGO, BCN, rGO-BCN_{800}, rGO-BCN_{900} and rGO-BCN_{1000}.

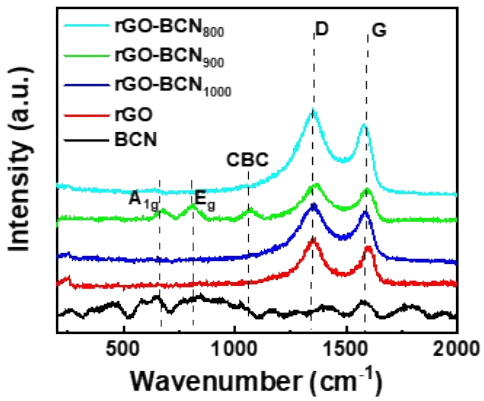

Fig. S5 Raman spectra of rGO, BCN, rGO-BCN_{800}, rGO-BCN_{900} and rGO-BCN_{1000}.

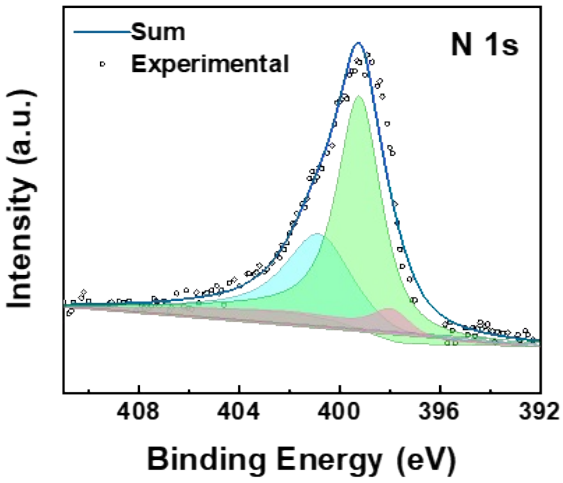

Fig. S6 N 1s spectra of rGO-BCN.

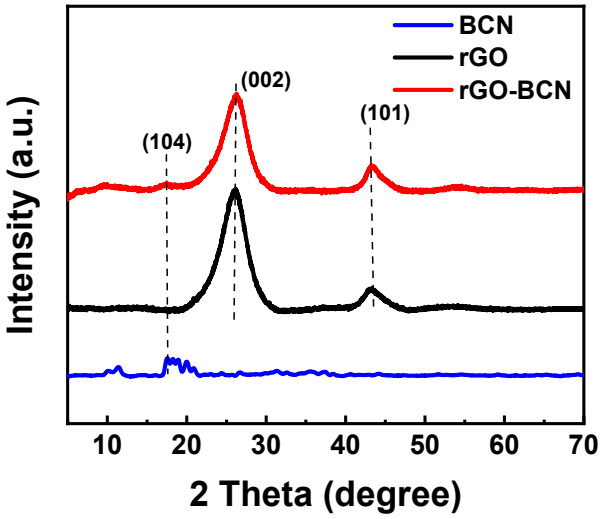

Fig. S7 XRD patterns of rGO, BCN and rGO-BCN.

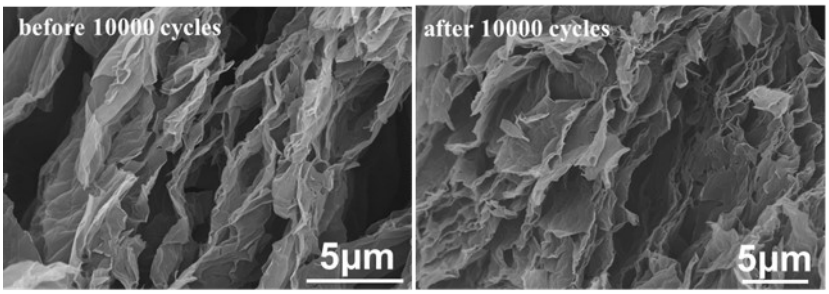

Fig. S8 SEM images of rGO-BCN before and after 10,000 cycles.

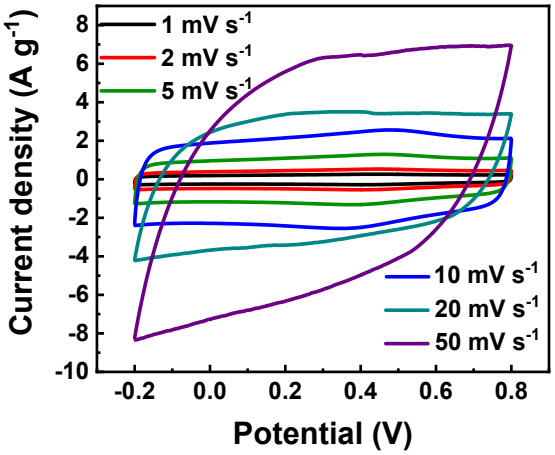

Fig. S9 CV curves of the rGO-$BCN_{1/4}$ fiber membrane at scan rates from 1 to 50 mV s^{-1}.

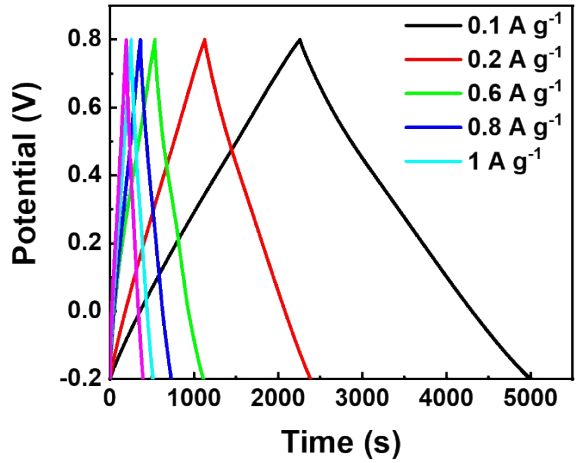

Fig. S10 GCD curves of rGO-$BCN_{1/4}$ fiber membrane at the current density from 0.1 to 1 A g^{-1}.

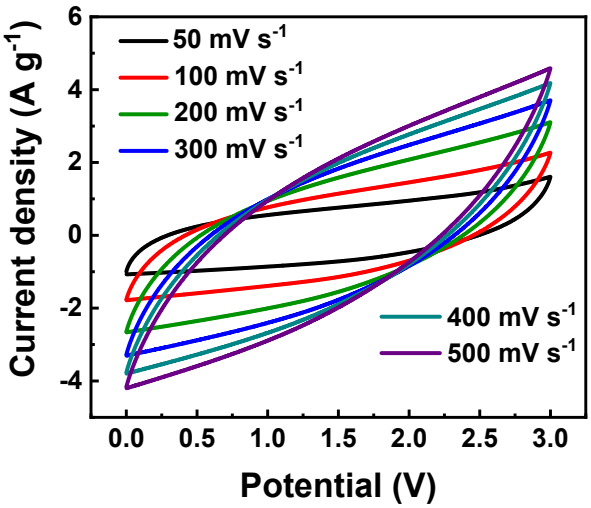

Fig. S11 CV curves of the FSCs at scan rate from 50 to 500 mV s^{-1}.

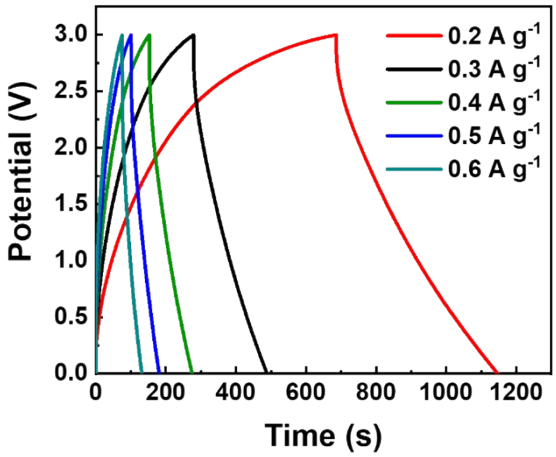

Fig. S12 GCD curves of FSCs at the current density from 0.2 to 0.6 A g^{-1}.

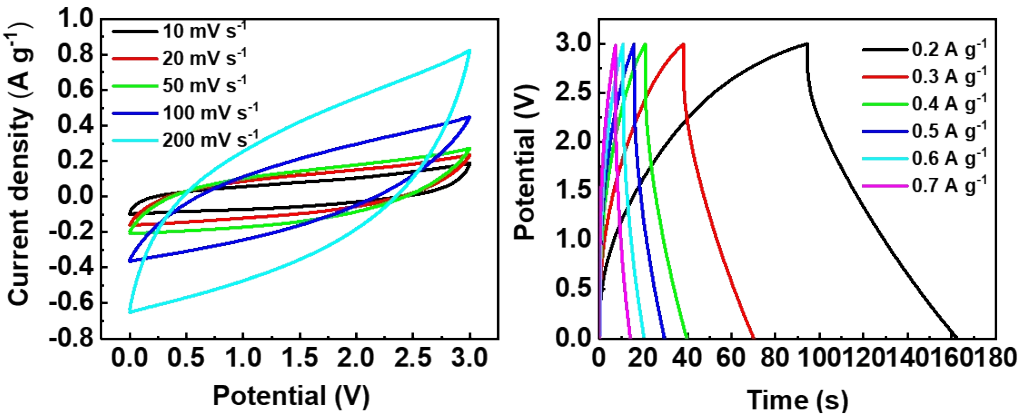

Fig. S13 CV curves of the FSC of rGO at scan rate of 10-200 mV s^{-1} and GCD curves of the FSC of rGO at the current density of 0.2-0.7 A g^{-1}.