

Electronic Supplementary Information for

**A bottom-up approach to design wearable and stretchable smart
fibers with organic vapor sensing behaviors and energy storage
properties**

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S1 Experimental

S1.0 Materials

Few-layer graphene (G-100) was purchased from Shanghai Simbatt Energy Technology Co. Ltd, China. The total oxygen content was 7.0-7.5 wt%, and the conductivity was about 700-1500 S m⁻¹. This material is denoted FLG unless otherwise defined. SBS D1102K (Kraton, USA) with butadiene/styrene molar ratio of 72/28 and a density of 0.94 g cm⁻³ was used as the polymer matrix. Sulphuric acid, acetone, chloroform, tetrahydrofuran, and cyclohexane were purchased from Sinopharm Chemical Reagent Co. Ltd., China. Poly(vinyl alcohol), PVA (Mw = 65,000 g mol⁻¹) used as the gel electrolyte and Polyvinylpyrrolidone, PVP (Mw=1,300,000) spinning helper polymer were purchased from Sigma-Aldrich. Carbon black (Super P conductive) was purchased from Alfa Aesar. All reagents were of analytical grade and used as received without further purification.

S1.1 Samples preparation

Preparation of fiber organic vapor sensor samples. FLG was dispersed in tetrahydrofuran (THF, 20 mL) and kept in an ultrasound bath for 4 h. Then, SBS was added in FLG/THF suspension (the SBS-G loading was fixed at 15 wt. %). The solution was agitated at 25 °C for 24 h and finally ultrasonicated for another 1 h at an output power of 45 W using a digital ultrasonic processor (S-450D-1/2, Branson) in a water bath prior to fiber spinning. The SBS-G fibers were fabricated through a facile and scalable wet-spinning process. Briefly, the spinning solutions were injected into a rotating ethanol coagulation bath using a 10-mL syringe with a 23-gauge hollow needle (as spinneret). The resulting fibers were drawn out vertically and wound onto a spool.

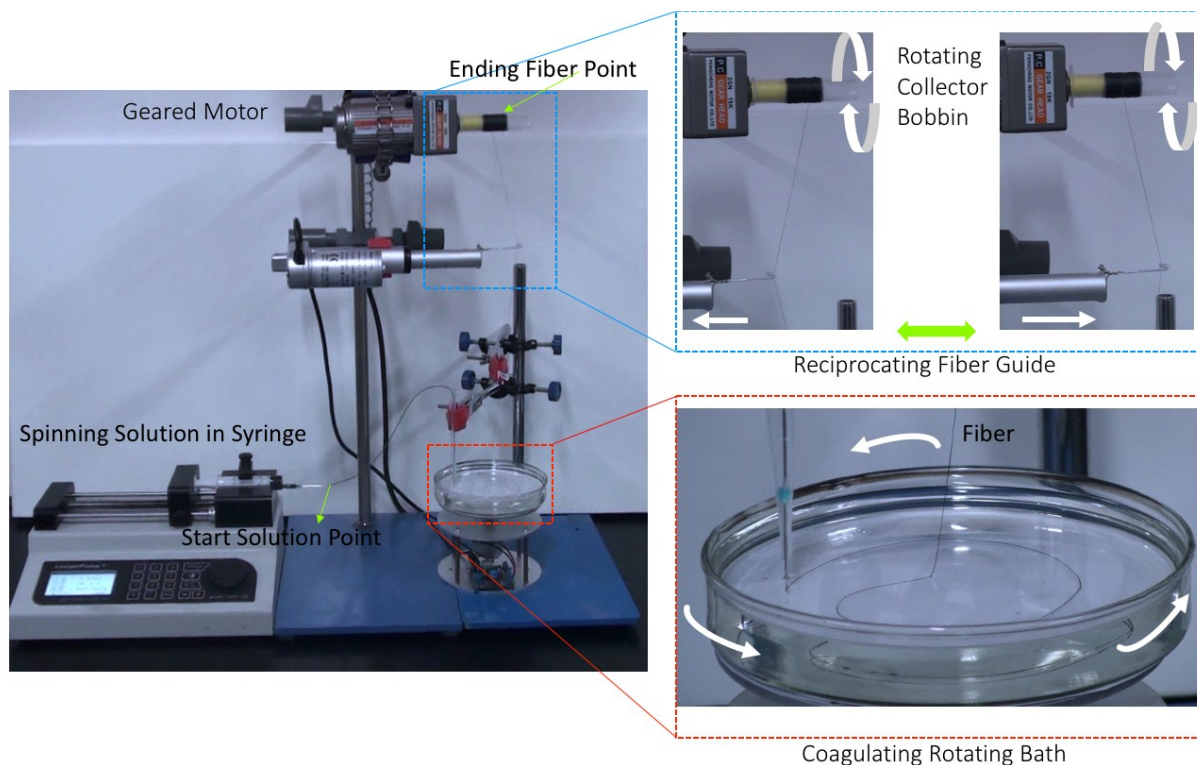


Figure. S1. Photographic image showing the process of fabricating the SBS-G composite fibers

Preparation of SBS-G nanofibers-coated electrodes samples. The fiber electrodes were prepared using SBS-7G fiber with the best conductivity. The SBS-7G fiber was modified via modified electrospinning technique which involved coating nanofibers of CB electrospinning solution as previously described in our work.¹ Briefly, the technique involved self-assembly of CB nanofibers onto SBS-7G fiber with the help of our homemade electrospinning machine previously described in ref². The spinning solution recipe was made by introducing 1.83wt/v% CB solution (made with distilled water: ethanol (7:4) solution) to 10 wt/v% PVP solution (made with ethanol/DMF solution). Then, a 5 mL portion of the spinning solution was added to 10 mL plastic

syringe for electrospinning. The alternating spinning nozzle speed was kept at 200 mm min^{-1} , with the SBS-7G fiber placed at a distance of 5 cm from the spinning nozzle. The solution flow rate to the spinning region was kept at $5.33 \text{ } \mu\text{l min}^{-1}$ and the voltage applied was $\sim 16 \text{ kV}$.

Preparation of SBS-G CB slurry-coated fiber electrodes samples. The SBS-7G fiber was immersed into the CB spinning solution and then withdrawn out and dried overnight at RT prior to characterization.

Preparation of the Gel Electrolyte. PVA 6 g was added to 60 mL of deionized hot water ($85 \text{ } ^\circ\text{C}$) under magnetic stirring. Then during the cool-down, 6 g of sulphuric acid was also added to PVA solution still under stirring. The gel electrolyte solution was left to cool to room temperature (RT) before use

Preparation of Devices. The devices were assembled as previously reported in our work¹ but with few changes. Briefly, the fiber samples employed were cut 2.5 cm and then assembled in the parallel-close distance (2 cm) with each other, and extra 0.5 cm left out which was further used as the terminals when conducting Silver paste was applied. Finally, to build a full device PVA/ H_2SO_4 gel electrolyte was introduced. The weight of the fiber electrodes lied around (average of 4 fibers) ~ 0.0825 , ~ 0.1275 and $\sim 0.130 \text{ mg cm}^{-1}$ for SBS- 7G, SBS-7G@CB and SBS-7G NCY, respectively. The active length was taken to be 2 cm.

S 1.2 Morphological and electrical characterization

The surface and cross-sectional (fractured in liquid nitrogen) morphologies of fibers were characterized using a scanning electron microscopy JEOL JSM-4800LV. The samples for SEM observation were sputter coated with platinum. The electrical conductivities of the fibers in the dry state were measured by a two-point probe method, in which probes were connected to a Keithley

2000 source meter. The electrical conductivity of the fiber was calculated using **Equation. S1**, where σ is the electrical conductivity (S cm^{-1}), R is the electrical resistance (Ω), and L and S are the length (cm) and cross-section area (cm^2) of the composite fiber, respectively.

$$\sigma = \frac{L}{R \times S} \quad (\text{S1})$$

The variation of electrical resistance was monitored online using a Keithley 2000 digital multimeter. The corresponding responsivity was calculated by the relative resistance change $((R - R_0)/R_0 \times 100\%$, where R_0 is the initial resistance of the sample in dry air and R is the real-time resistance measured during the experiment upon the alternating exposure to various analytes and dry air).

The fiber mechanical properties were probed using Instron 5996 universal testing machine. For tensile tests, the fibers were stretched to 400% at a strain rate of 20 mm min^{-1} . The elastic recovery, ER_ε at each applied strain, ε was calculated using **Equation. S2** and ε_R is the residual strain after stretching-releasing tests (**Figure. S5b**).

$$ER_\varepsilon = \frac{\varepsilon - \varepsilon_R}{\varepsilon} \cdot 100 \quad (\text{S2})$$

CV, GCD, and EIS tests of the devices were done on an electrochemical workstation (CHI 660E) using a two-electrode system. EIS parameters were 5 mV ac amplitude and frequency range 1MHz–100 kHz. The capacitance (C) of the single yarn electrode can be obtained from the CV curves and GCD curves according to Equation (S3) and (S4), respectively as follows:

$$C = \frac{\int I(V)dV}{v\Delta V} \quad (S3)$$

$$C = \frac{I}{dV/dT} \quad (S4)$$

Then, single-electrode specific capacitance (C_{sp}) can be calculated using Equation (S5)

$$C_{sp} = \frac{2C}{X} \quad (S5)$$

The volumetric energy density (E) and the areal power density (P) were calculated based on Equation (S6) and (S7), respectively as follows:

$$E = \frac{1}{7200} C_{Dev} (\Delta V)^2 \quad (S6)$$

6)

$$P = \frac{E_D}{\Delta t} 3600 \quad (S7)$$

Where C_{Dev} represents the device capacitance, v is the potential scan rate ($V s^{-1}$); ΔV is the potential window (V); I (A) is the constant discharging current, and dT is the discharging time (s).

X is either the mass (g) or volume, V (cm^3) or area, A (cm^2) of the yarn which can be calculated as follows in Equations (S8 and S9)

$$V = \pi D^2 L / 4 \quad (\text{S8})$$

$$A = \pi D L \quad (\text{S9})$$

Where D (cm) is the diameter of the fiber with length L (cm)

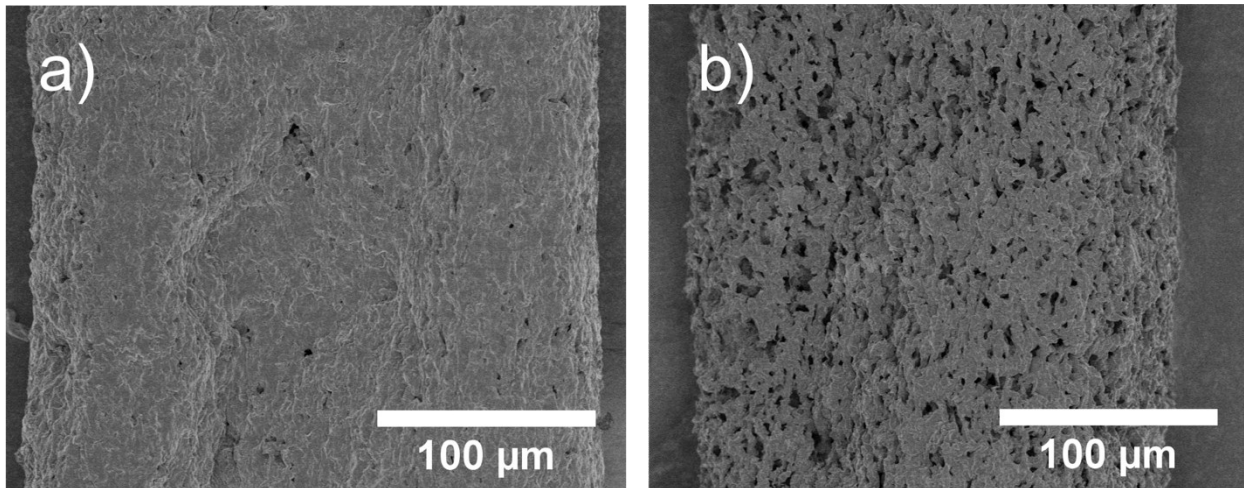


Figure. S2. SEM surface images of SBS-G composite fibers. (a) SBS-5G fiber and (b) surface image of the SBS-7G fiber.

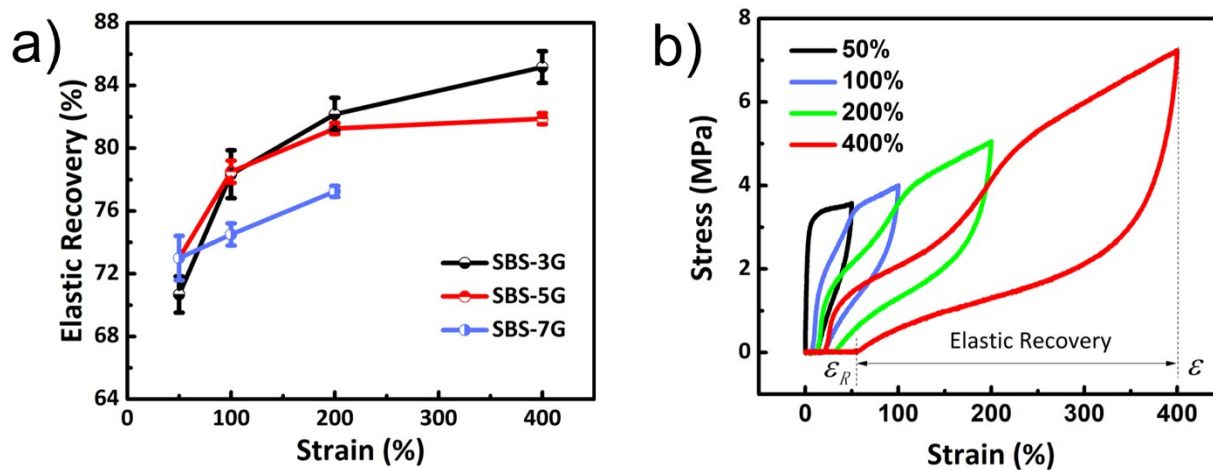


Figure. S3. (a) The elastic recovery of SBS-G during cyclic stretching-releasing analysis as a function of strain and (b) cyclic elastic recovery performance curves of SBS-3G fibers for with growing strain ranging from 50 to 400%.

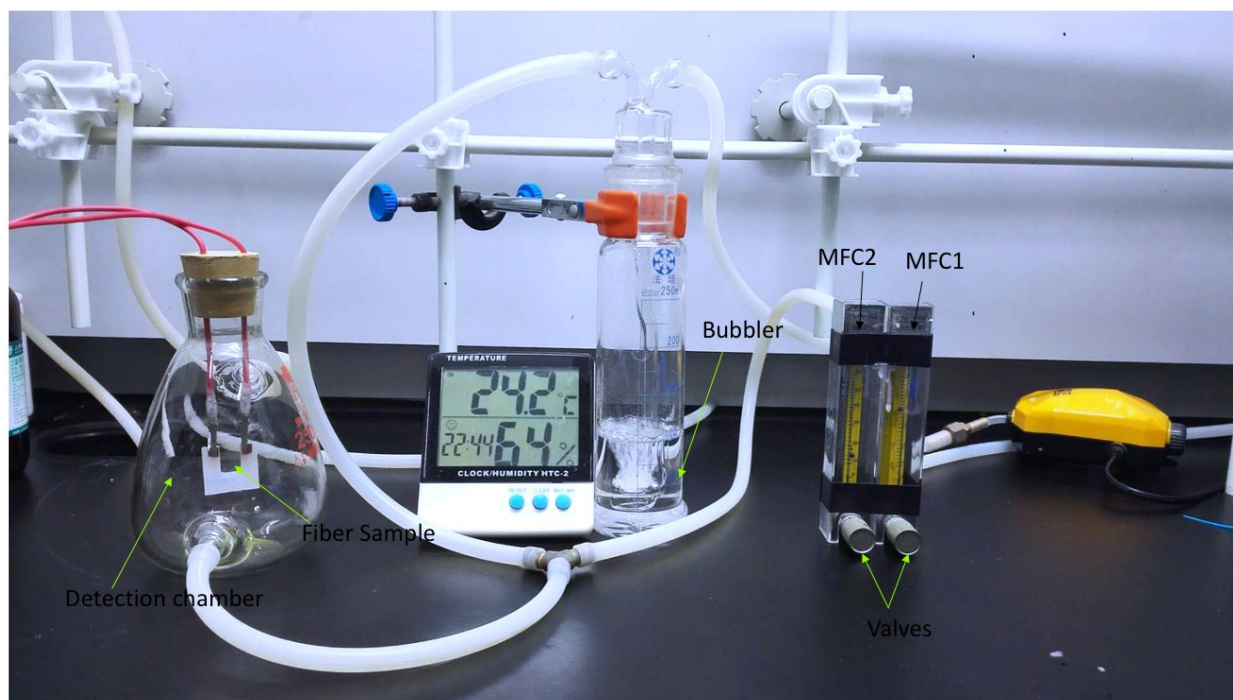


Figure. S4. Photographic image showing the experimental setup for the detection of the vapor sensing behavior of SBS-G composite fibers.

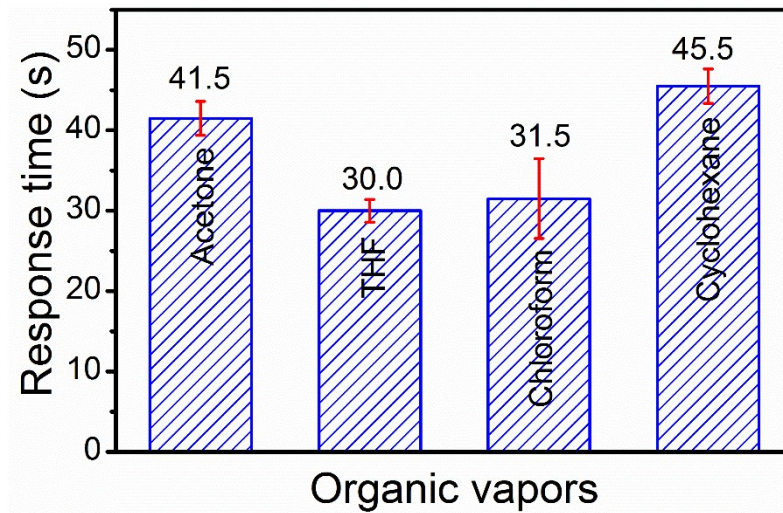


Figure. S5. The Response time of SBS-3G fibers towards various VOC vapors.

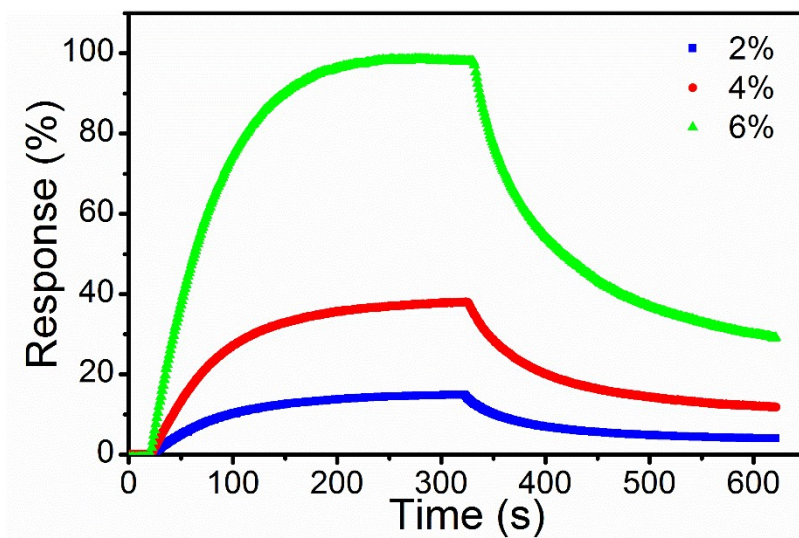


Figure. S6. Responses of SBS-G composite fibers upon exposure to different concentrations of cyclohexane vapor.

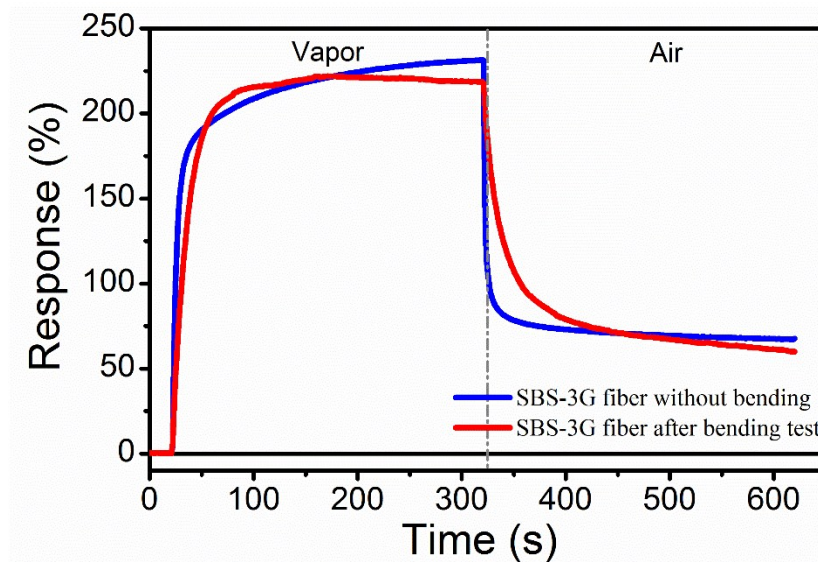


Figure. S7. Relative resistance changes of SBS-3G composite fiber without bending and after 1000 bending-straightening cycles upon cyclic exposures to 10% cyclohexane vapor.

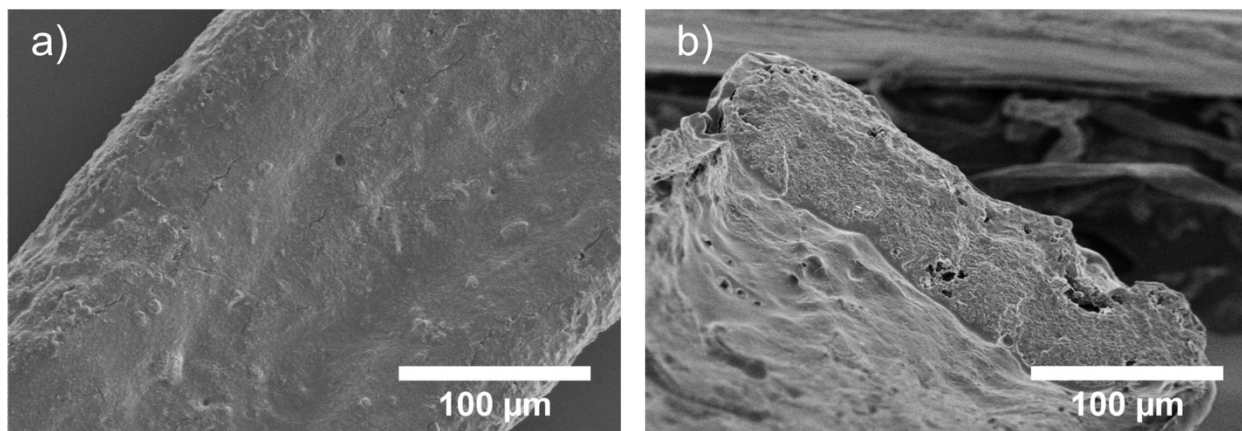


Figure. S8. SEM images of SBS-7G@CB. (a) surface and (b) cross-section.

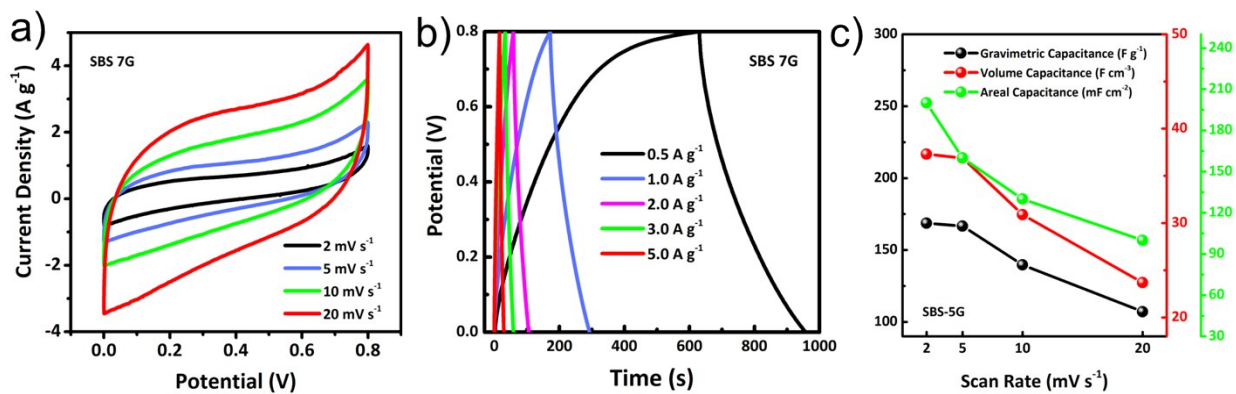


Figure. S9. Electrochemical performance of the Sneat SBS-7G device. (a) CV and (b) GCD curves at various scan rates and current density, respectively, (c) variation of specific capacitance values of the device with scan rate from obtained from CV curves in (a).

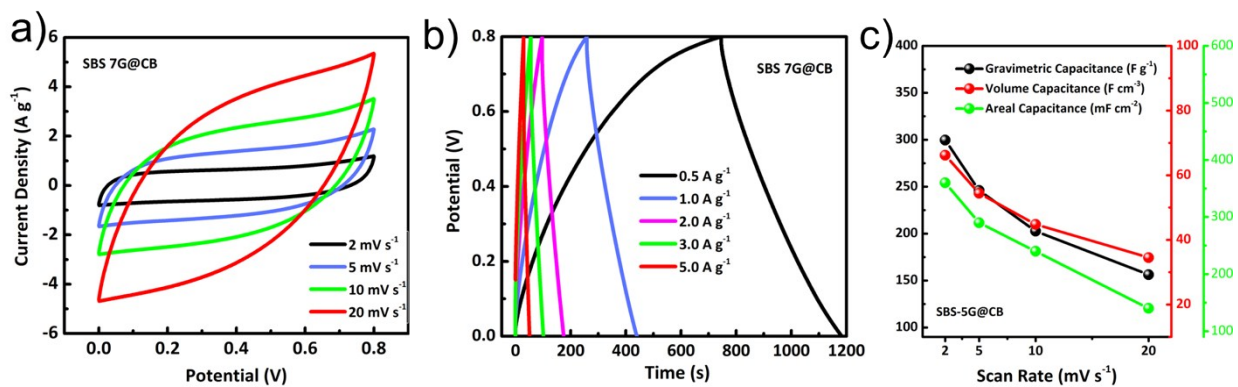


Figure. S10. Electrochemical performance of SBS-7G@CB device. (a) CV and (b) GCD curves at various scan rates and current density, respectively, (c) variation of specific capacitance values of the device with scan rate from obtained from CV curves in (a).

Table. S1. Variation of the energy and power density of the SBS-7G NCY device with the current density

Current Density (A g⁻¹)	Energy Density (mWh cm⁻³)	Power Density (mW cm⁻³)
0.5	6.6	49
1	6.0	100
2	5.3	229
3	4.8	573
5	4.0	692

Supporting information References

1. M. Tebyetekerwa, Z. Xu, W. Li, X. Wang, I. Marriam, S. Peng, S. Ramkrishna, S. Yang and M. Zhu, *ACS Applied Energy Materials*, 2018, **1**, 377-386.
2. M. Tebyetekerwa, X. Wang, Y. Wu, S. Yang, M. Zhu and S. Ramakrishna, *J. Mater. Chem. A*, 2017, **5**, 21114-21121.