Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.


# Supplementary Information

## Fiber-reinforced Monolithic Supercapacitor with Interdigitated Interfaces

Fanshu Yuan,[1,2§] Devashish Salpekar,[1§] Abhijit Baburaj[1§], Anand B. Puthirath[1], Sakib Hassan[3], Francisco C Robles Hernández[4], Hossein Robatjazi[3], M.A.S.R. Saadi[1], Soumyabrata Roy[1], Dongping Sun[2*], Nicholas A. Kotov[5*], Muhammad M. Rahman[1*], and Pulickel M. Ajayan[1*]

[1]Department of Materials Science and NanoEngineering, Rice University, TX 77005, USA
[2]Chemicobiology and Functional Materials Institute, Nanjing University of Science and Technology, Nanjing 210094, China
[3]Department of Electrical and Computer Engineering, Rice University, TX 77005, USA
[4]Department of Materials Science and Engineering, University of Houston, TX 77204, USA
[5]Department of Mechanical Engineering, University of Michigan, Ann Arbor, MI, USA

*Correspondence to: sundpe301@163.com, kotov@umich.edu, mr64@rice.edu, ajayan@rice.edu

§ Equal contribution

**Electrochemical Calculations**

The specific capacitance of the electrode ($C_{sp}$) materials is calculated from Cyclic Voltammetry using the following equation:

$$C_{sp} = \int \frac{Idv}{M \bullet v \bullet dv} \qquad \text{(S1)}$$

Here, integration of current (*I) over the potential window (dv)* is equal to the area of CV curve and *M* is the mass of electrode material *(g)*, *v* is the scan rate (mV /sec). The capacitance from GCD was calculated using the following formula:

$$\textit{From GCD:} \quad C_{sp} = \frac{Idt}{M \bullet dv} \qquad \text{(S2)}$$

Where *I* is the applied current, *dv* potential window and *dt* discharge time (s) and *M, the* mass of active material (g). The areal capacitance of the electrodes is derived by replacing the mass by the area (A) in.

$$\text{From GCD:} \quad C_{\text{Areal}} = \frac{Idt}{A \bullet dv} \quad \text{(S3)}$$

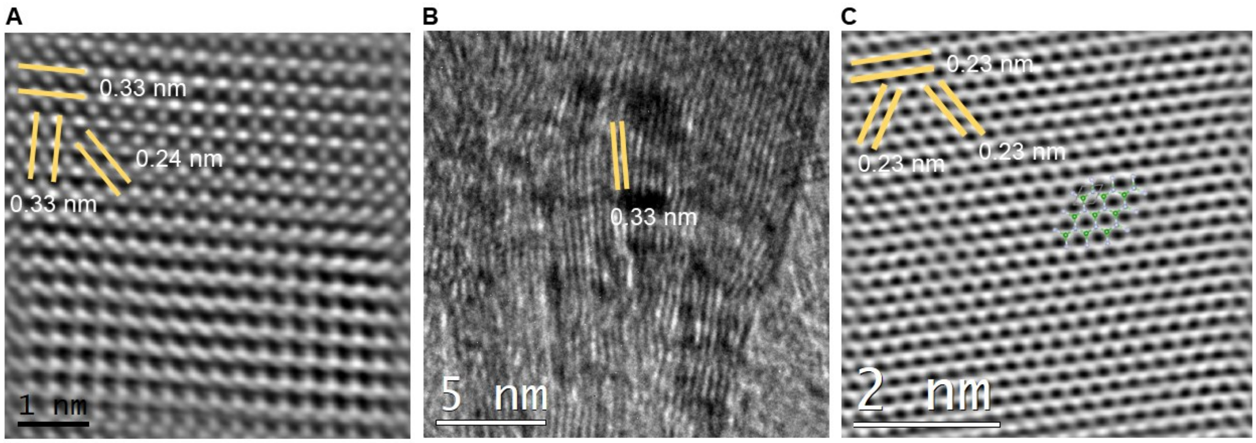


**Figure S1.** HRTEM analysis of pure (A) PANFs, (B) GE from GE-PANF and (C) BNNS from BNNS-PANF

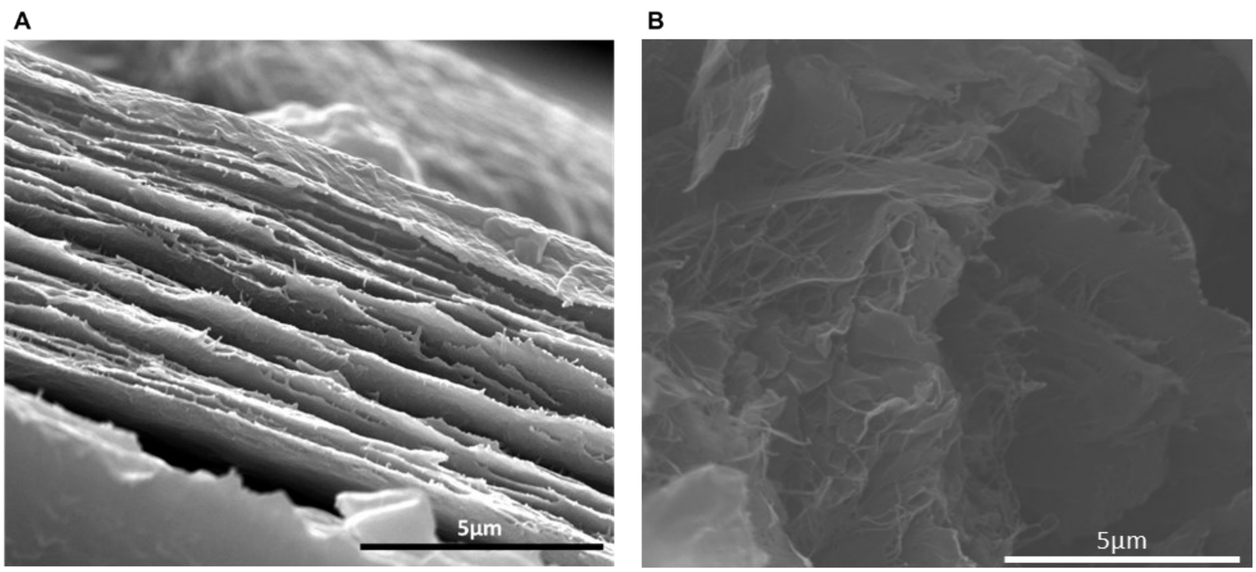


**Figure S2.** Scanning electron microscopic images of the BNNS-PANF separator (A) and GE-PANF (B).

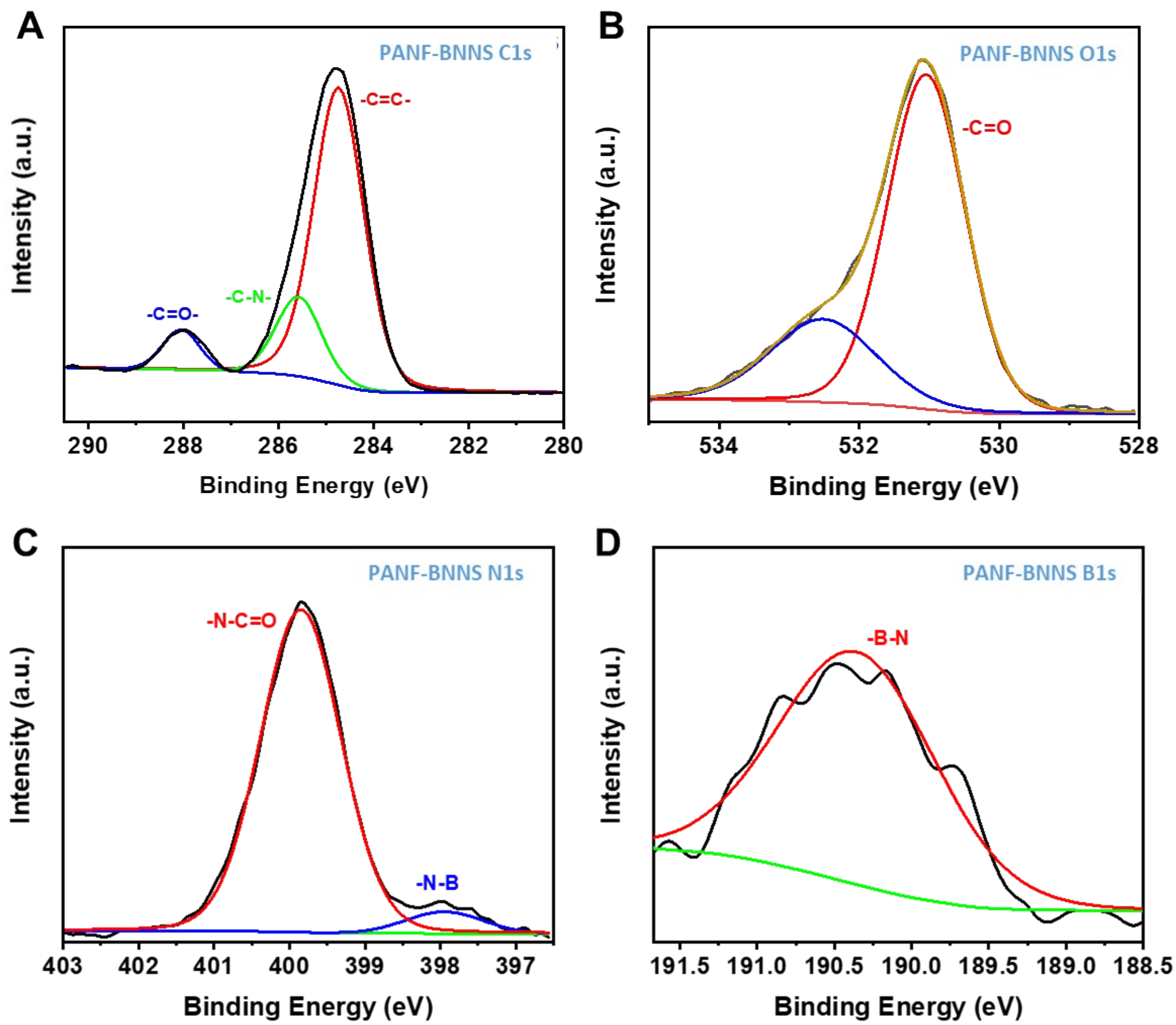


**Figure S3.** Deconvoluted spectrum of carbon (A), oxygen (B), nitrogen (C) and boron (D), from the BNNS-PANF separator.

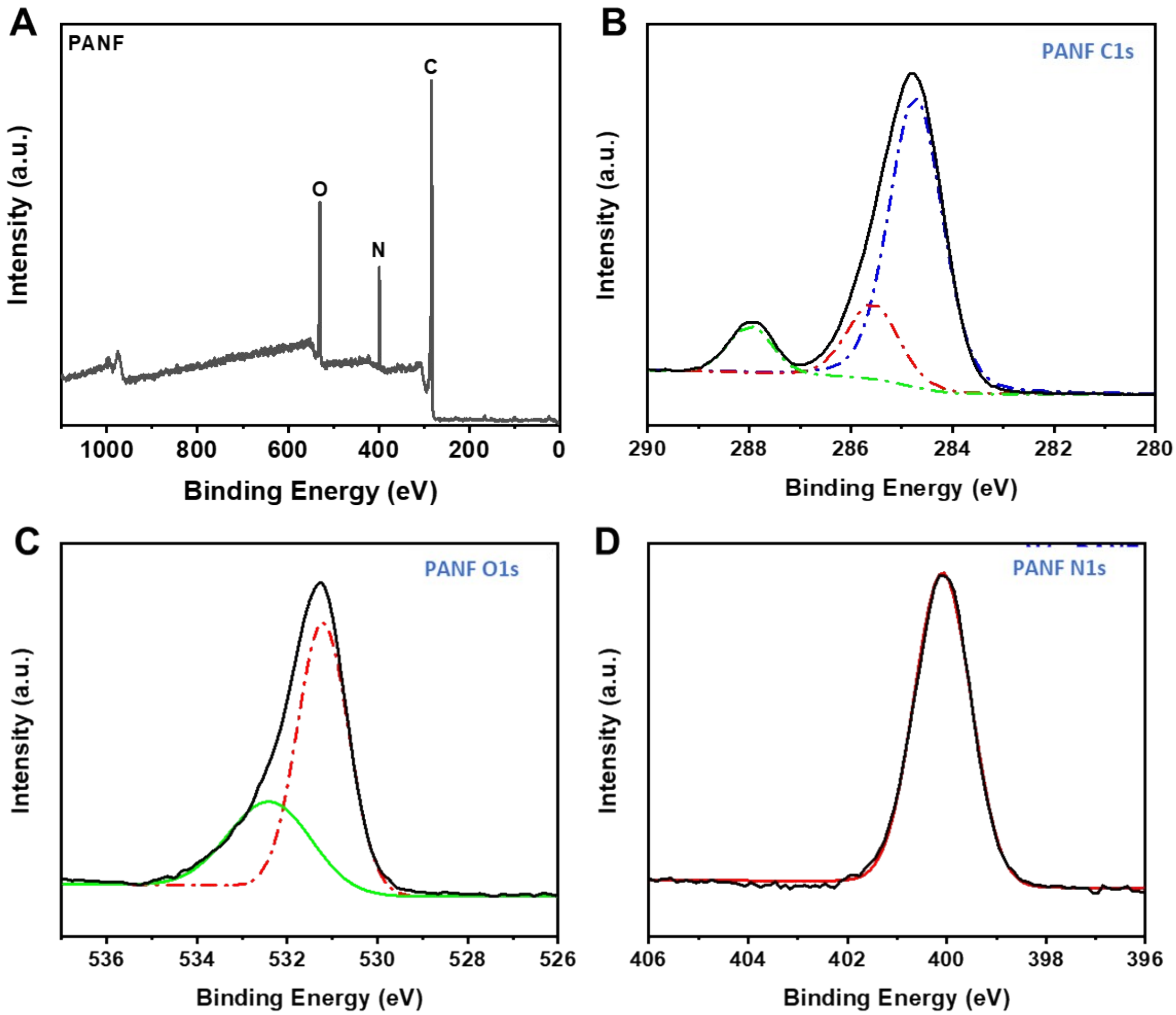


**Figure S4.** Survey scan for the PANF (A). Deconvoluted analysis of the carbon (B), oxygen (C) and nitrogen (D) from PANF.

C1s spectra of PANF was fitted with three peaks. The first peak originating at 284.5eV is assigned to the $sp^2$ hybridized carbon atoms in the aromatic rings of PANF. The second peak at 285.4eV corresponds to the carbon atoms bonded to nitrogen in the aramid structure. The final peak 287.9eV was assigned to the -C=O carbon atoms in the PANF structures. The ratio of the three area under the curves is close to 4:2:1, which matches with the exact structure of PANFs.

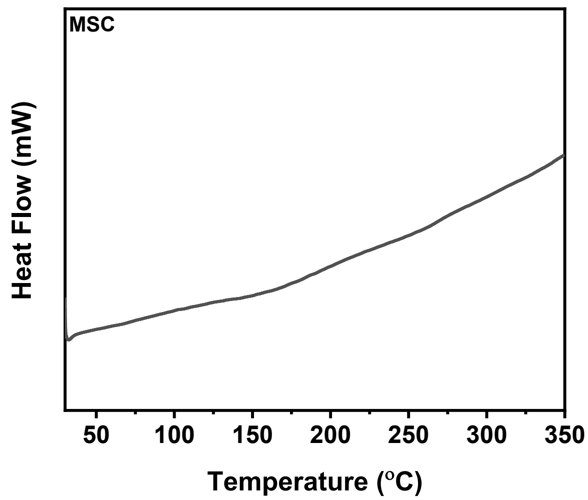


**Figure S5.** Differential Scanning Calorimetry of the monolith supercapacitor.

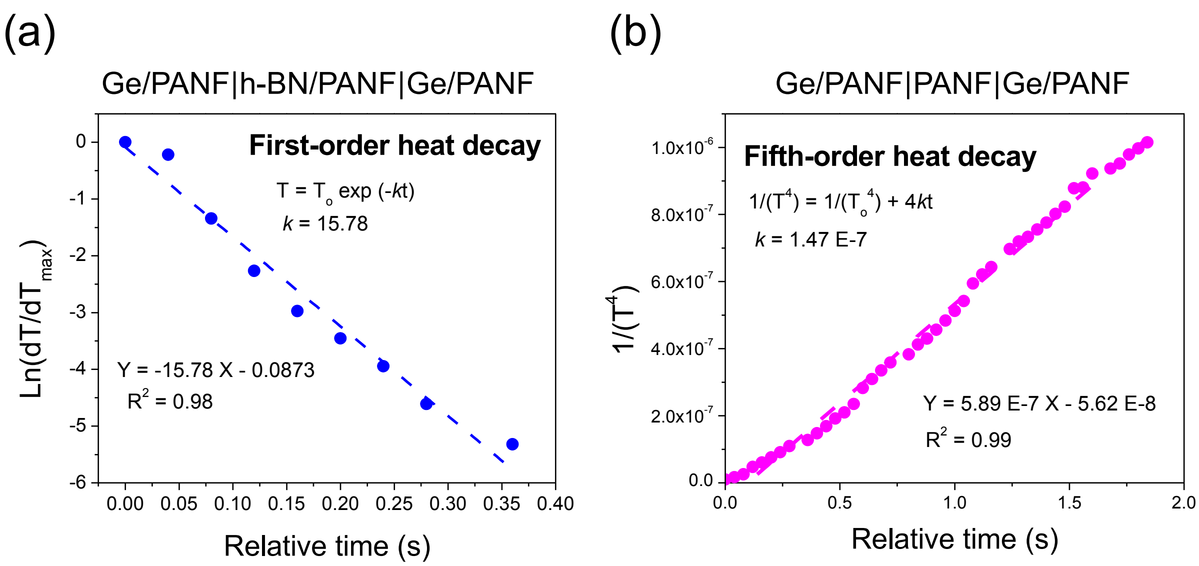


**Figure S6.** Measurements of heat dissipation rate. The rate of heat dissipations for the h-BN modified supercapacitor (a) and a non-modified supercapacitor (b) were calculated from fitting the temperature, T values (after the light was turned off) into a first-order and fifth-order decay, respectively. The obtained rate constants ($K$) for the heat dissipations were 15.78 and 1.47*$10^{-7}$ for the h-BN modified and non-modified samples, respectively. The initial surface temperature in each case was 105 $^{o}$C (See Fig. 3c of the main article for more details).

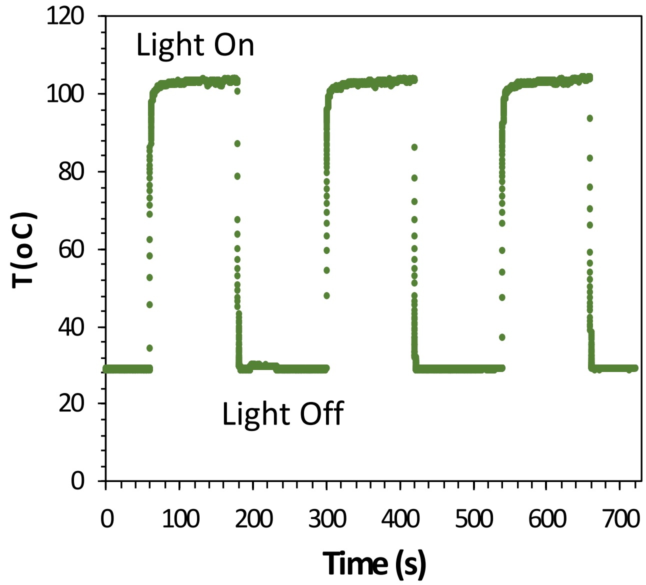


**Figure S7.** Reproducibility of the surface temperature measurements measured with the IR camera under multiple light on-off cycles. Measurements were performed on the h-BN modified supercapacitor. A CW (532 nm, 3 mm spot size) laser source at 40 mW illumination power was used for the irradiation.

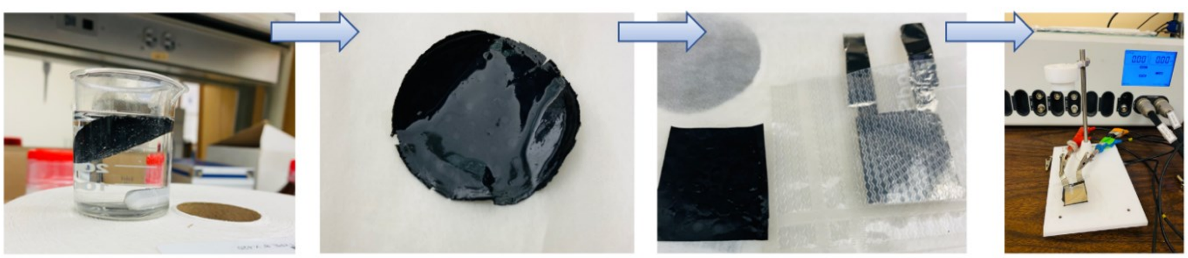

**Figure S8.** Ad-hoc pouch cell assembly of monolith supercapacitor for testing at various bending angles.

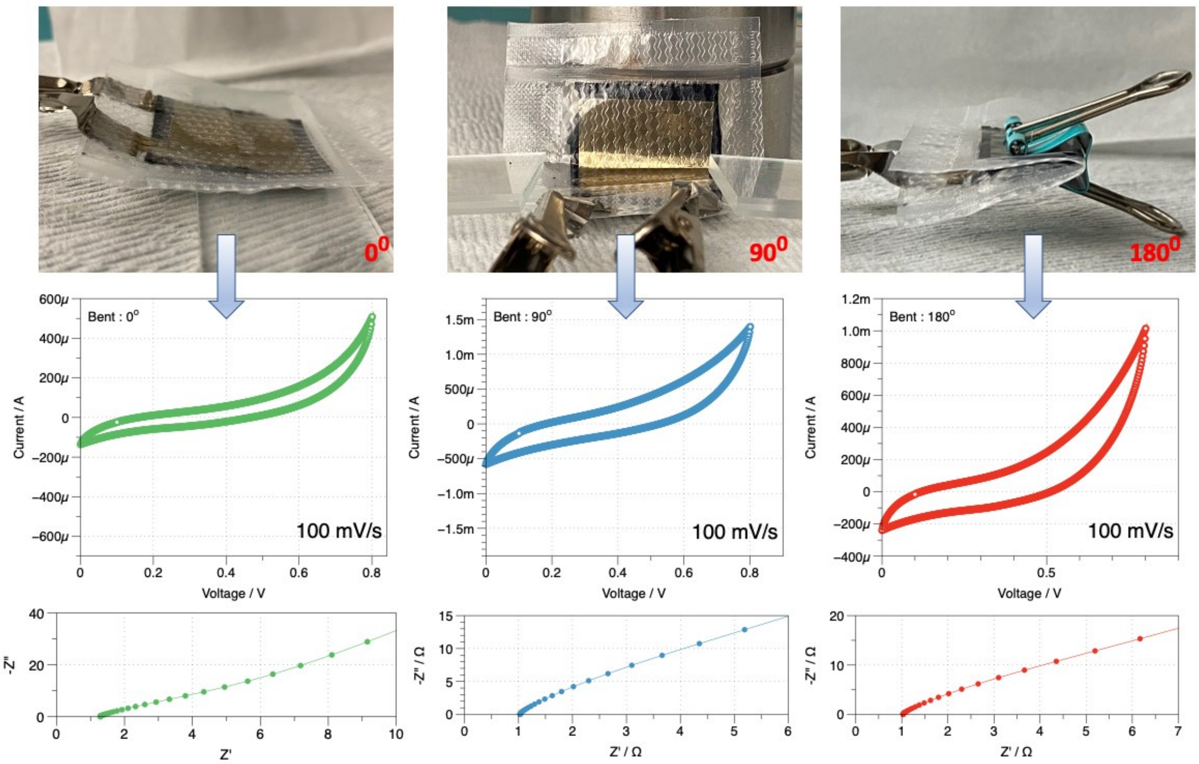


**Figure S9.** Electric Double Layer Capacitor (EDLC) and impedance behavior of MSC under different bend conditions.

We have conducted the study on electrochemical performance (EDLC) of the device under various deformations. We assembled an ad-hoc pouch cell to test the operability of the monolith supercapacitor under different bent conditions. We prepared a MSC with ~4 mm diameter and sliced a rectangular piece ( 2cm x 3cm) and assembled a supercapacitor after wetting the same in 6M KOH for 6 h and packaged it between two SS foils current collectors. The device was finally thermally sealed in a transparent insulating membrane. The various steps involved in the making pouch cell are shown in **Figure S8**. As a proof-of-concept, cyclic voltammetry and impedance spectroscopy were performed to test the EDLC behavior at different bent conditions, for instance, $0^o$, $90^o$ and $180^o$ using the ad-hoc pouch-cell. Being the pouch cell assemble is ad-hoc the CV curves are not square-like but more resistive-like. However, the EDLC capacitance (with resistance) behavior of the cell is observed and the same is retained even at $90^0$ and $180^0$ bent conditions (**Figure S9**).

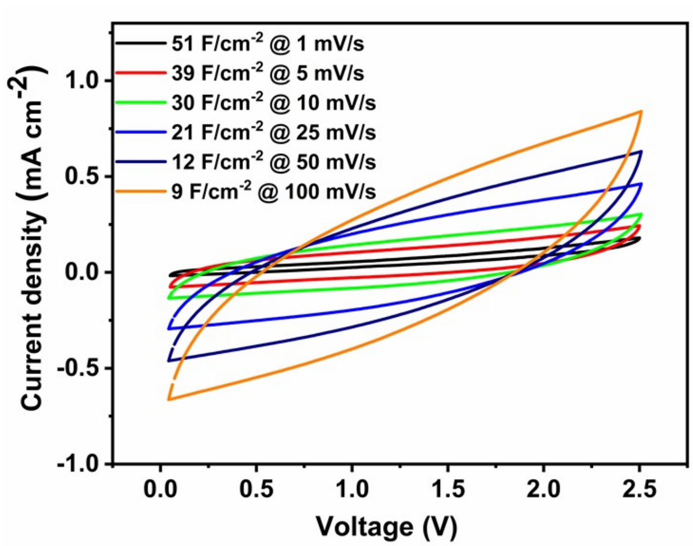


**Figure S10.** High-temperature cyclic voltammetry of the MSC with EMI-TFSI at 100 $^0$C.