Supporting Information

A Shape-Memory Supercapacitor Fiber

Jue Deng, Ye Zhang, Yang Zhao, Peining Chen, Xunliang Cheng, and Huisheng Peng*

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Supporting Information

Experimental section

CNT arrays were grown by chemical vapor deposition with Fe (1.2 nm)/Al_2O_3 (3 nm) on a silicon substrate as the catalyst at 740 ℃. Ethylene was used as the carbon source, and a gas mixture of Ar and H_2 was used as the carrier gas. The flow rates of Ar, H_2, and C_2H_4 were typically 400, 30, and 90 sccm, respectively. CNT arrays with thicknesses of about 250 μm were mainly used in this work. The CNT was multi-walled typically with a diameter of ~10 nm.

The structure was characterized by scanning electron microscopy (SEM) (Hitachi FE-SEM S-4800 operated at 1 kV) and transmission electron microscopy (JEOL JEM-2100F operated at 200 KV). The charging and discharging tests and cyclic voltammetry tests of the shape-memory supercapacitor were made by an electrochemical workstation (CHI 660D). The long-term cycle tests were taken by an Arbin electrochemical station (MSTAT-5 V/10 mA/16Ch). The heating of the shape-memory supercapacitor was realized by a Hot Plate (MODELKW-4AH). The elongating deformation was characterized by an HY0350 Table-top Universal Testing Instrument.
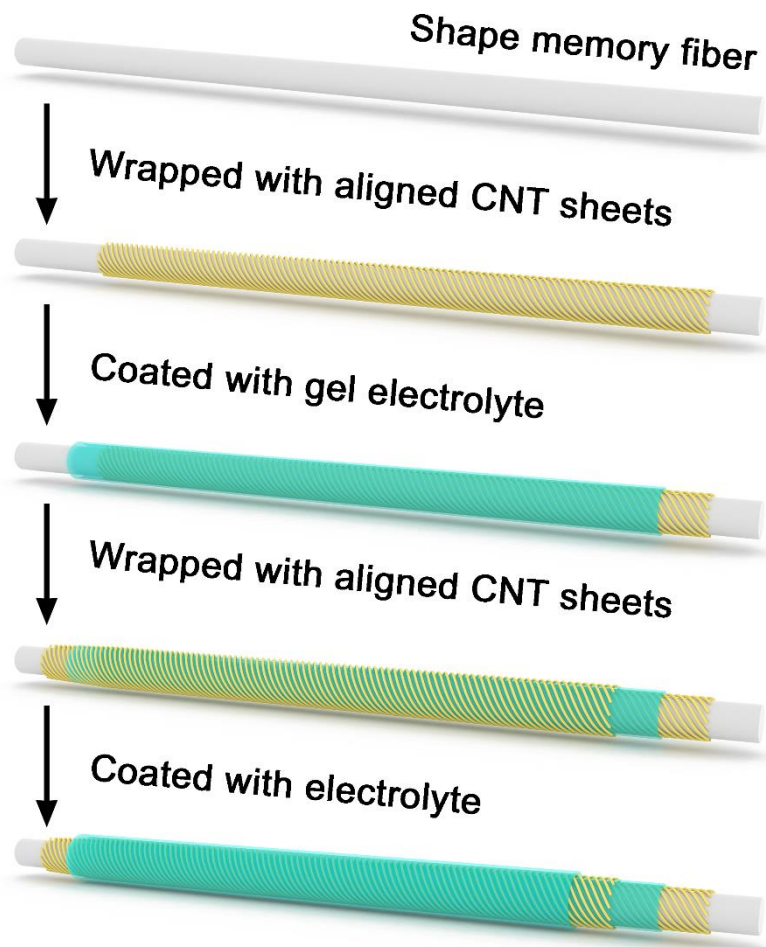
Shape memory fiber

Wrapped with aligned CNT sheets

Coated with gel electrolyte

Wrapped with aligned CNT sheets

Coated with electrolyte

Figure S1. Schematic illustration to the fabrication process of shape-memory, fiber-shaped supercapacitor (SFSC) with a coaxial structure.
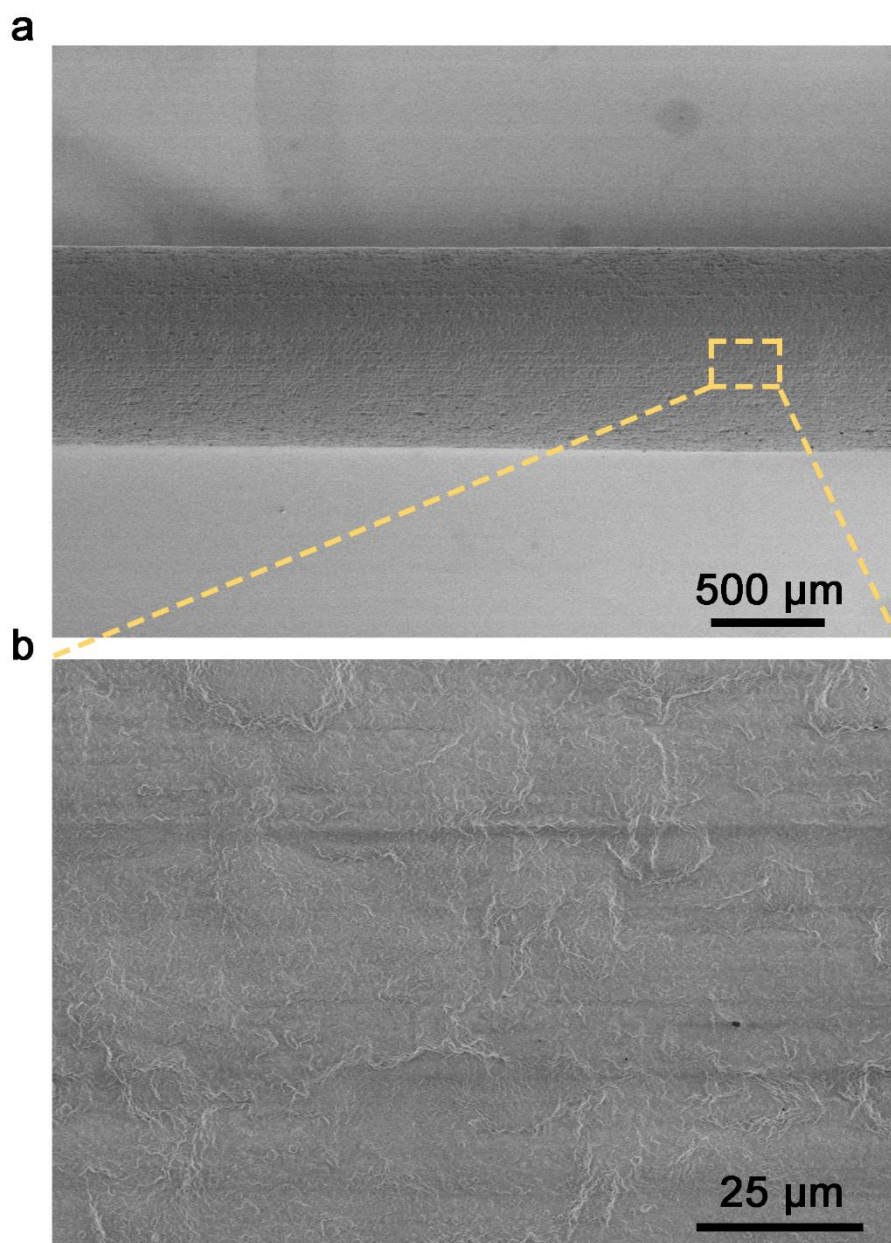
Figure S2. (**a**) Scanning electron microscopy (SEM) images of shape-memory polyurethane substrate. (**b**) Higher magnification of (**a**).
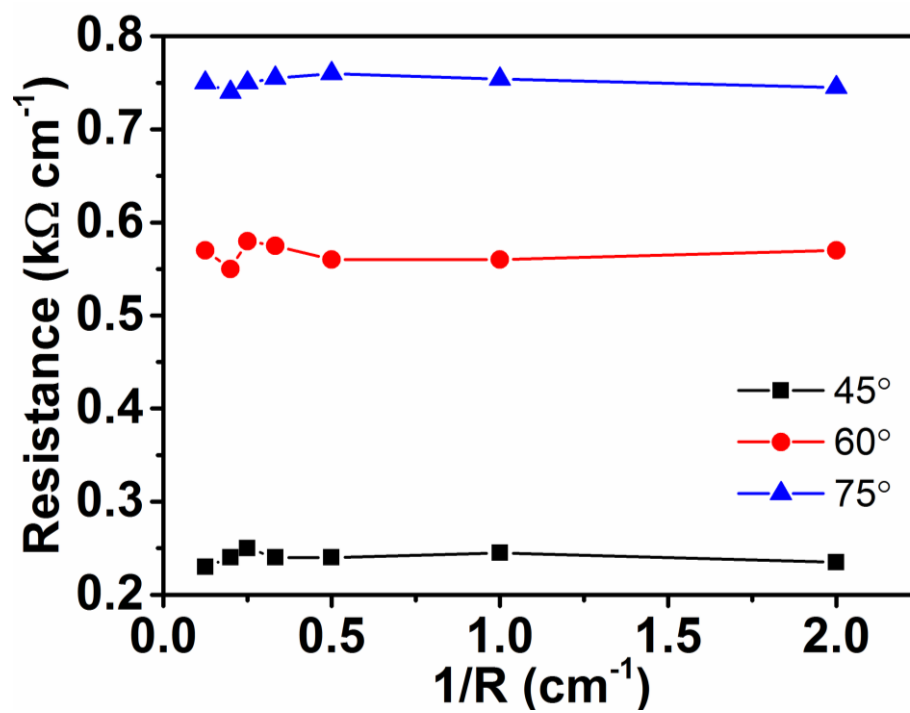
Figure S3. Dependence of electrical resistance for the SFSC electrode on the curvature of the flexural shape (thickness of the CNT, 560 nm).
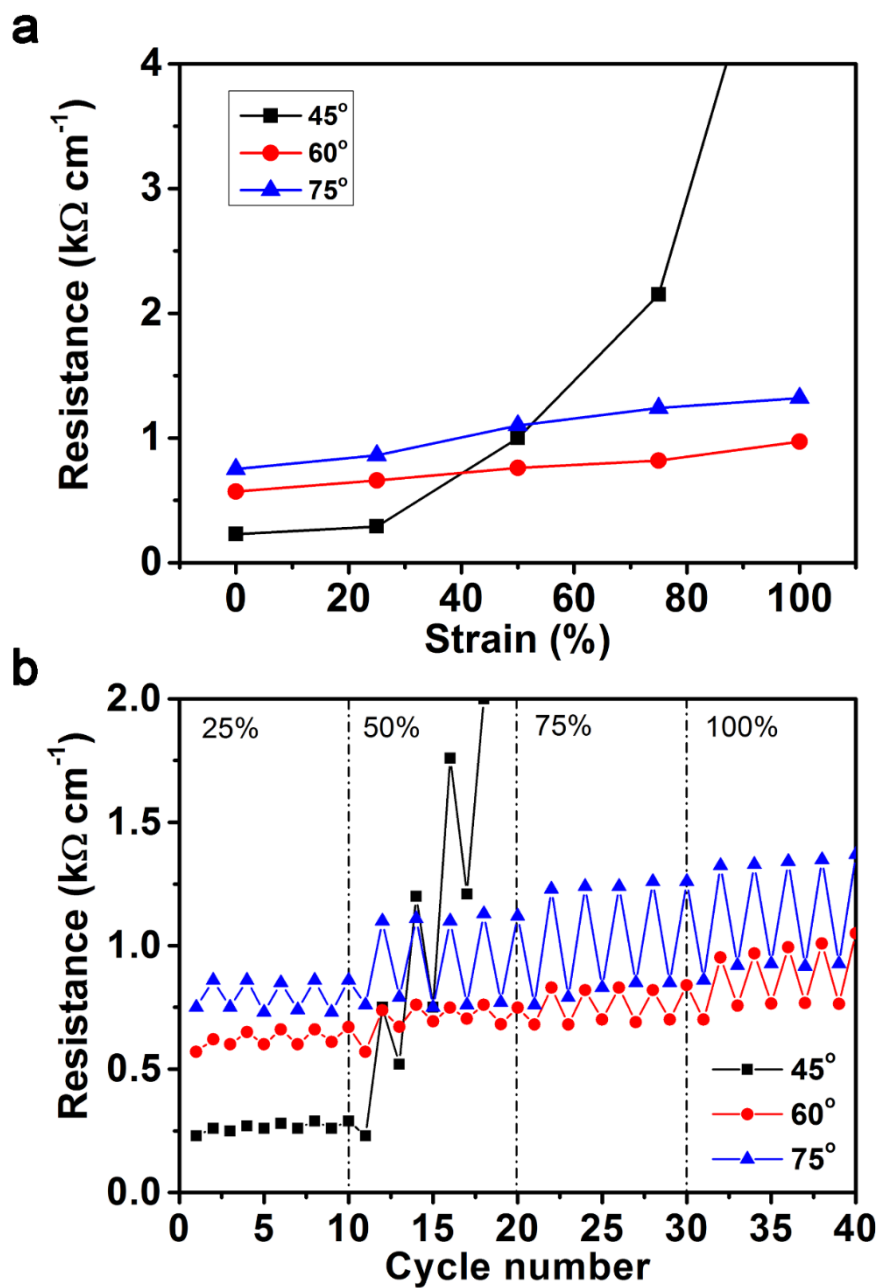
Figure S4. (**a**) Dependence of electrical resistance of the SFSC electrode on the strain at the elongated shape (thickness of the CNT, 560 nm). (**b**) Dependence of electrical resistance on the deformation and recovery cycle number.
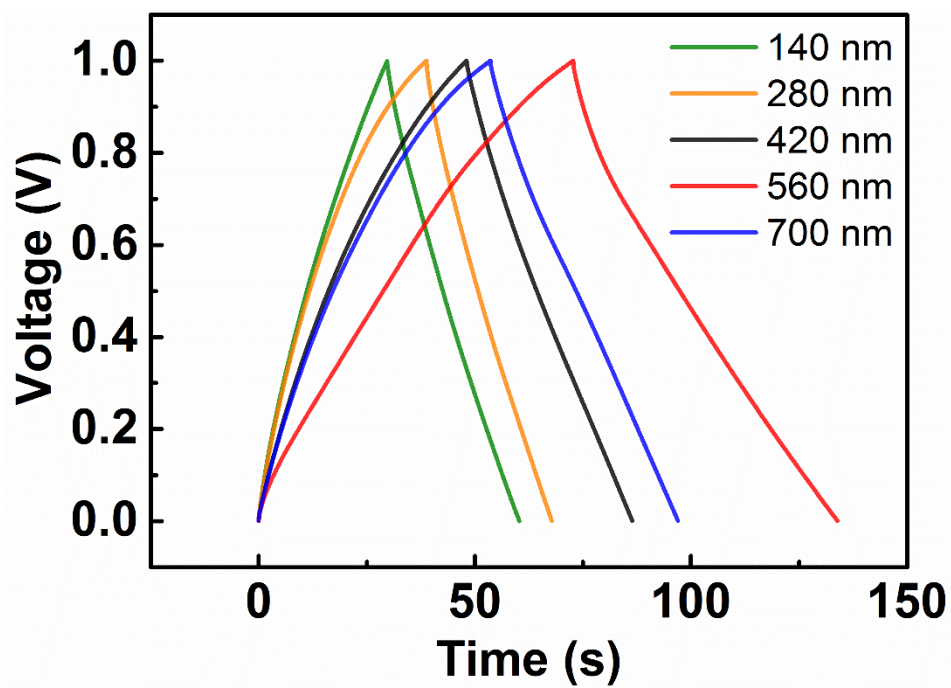
Figure S5. Galvanostatic charging and discharging curves of SFSCs with different thicknesses of CNT layers. The measurements were carried out at a current density of 0.1 A g^{-1}.
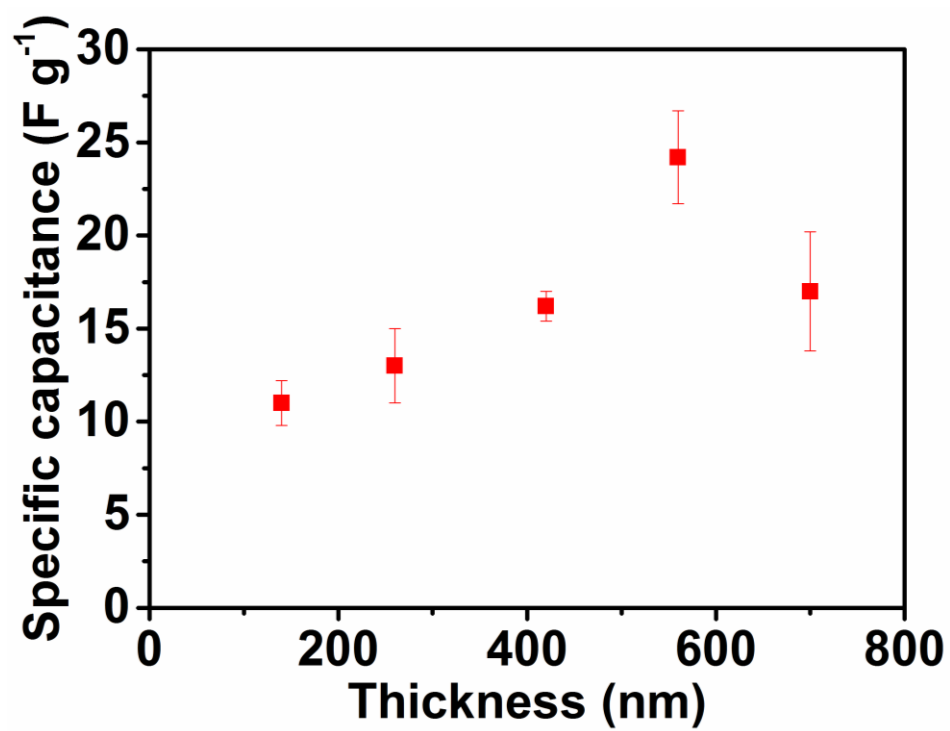
Figure S6. Dependence of specific capacitance on the thickness of CNT layer.
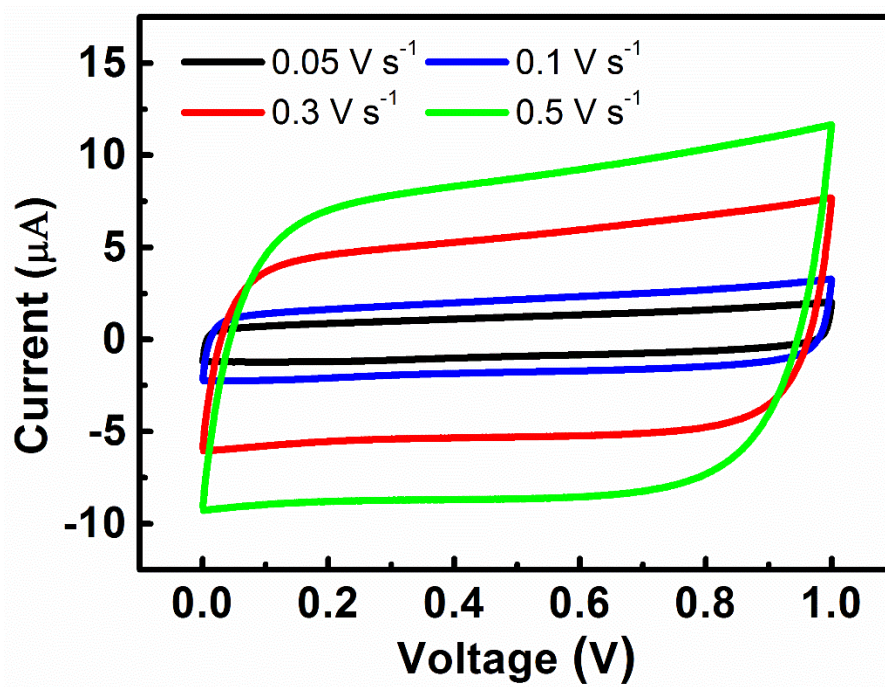
Figure S7. CV curves of an SFSC with the CNT thickness of 560 μm at increasing scan rates from 0.05 to 0.5 V s^{-1}.
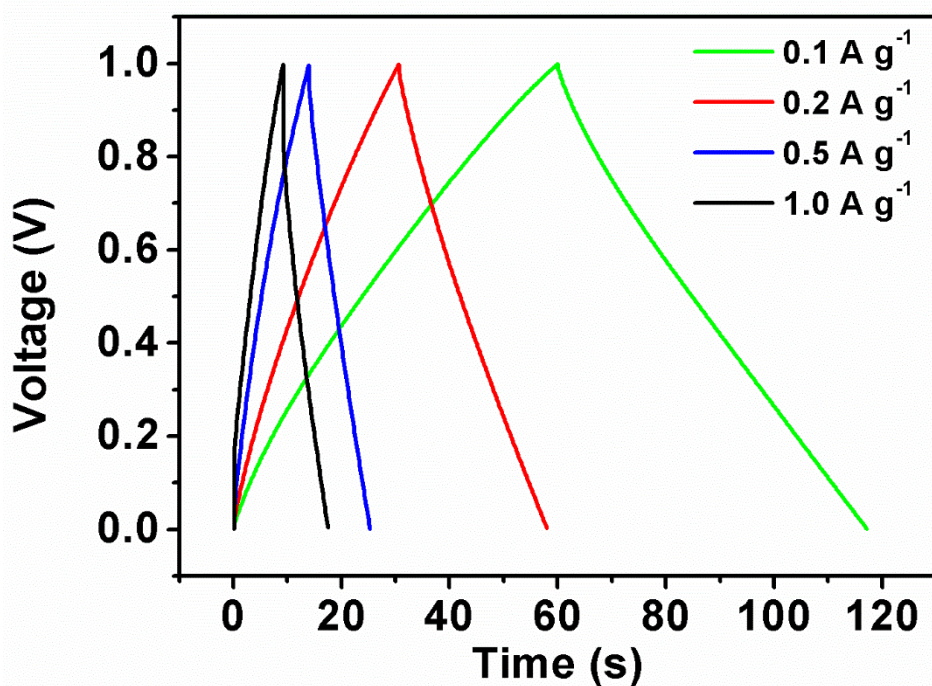
Figure S8. Galvanostatic charge-discharge curves of a fiber-shaped supercapacitor with the CNT thickness of 560 nm at increasing current densities from 0.1 to 1.0 A g^{-1}.
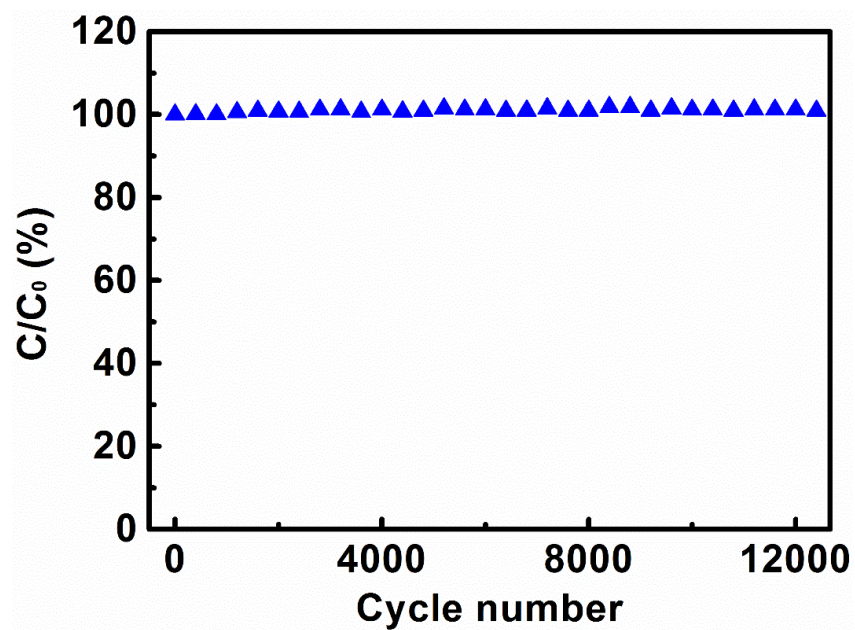
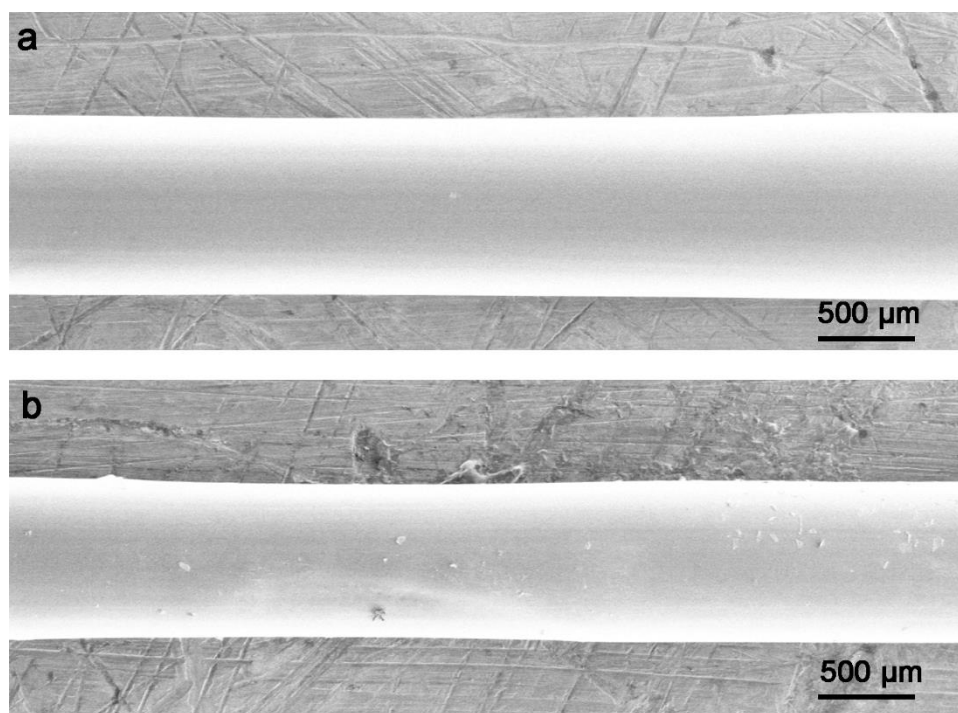**Figure S9**. Cyclic performance of the SFSC at a current density of 1 A g^{-1}.

Figure S10. SEM images of the surface of the fiber electrode with the electrolyte before (**a**) and after (**b**) deformation.