## Supporting Information

# Fiber-shaped Asymmetric Supercapacitors with Ultrahigh Energy

### **Density for Flexible/Wearable Energy Storage**

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#### 1. Experimental Section

ALD ZnO seed layer. Our purpose is grow aligned and ordered ZnO-NWs on CNT fiber using a wet chemical method. However, due to the deficiency of dangling bond on the surface of CNT fiber, it is difficulty to grown ZnO NWs on the surface of CNT fiber directly. First, the chemical modification of CNT fibers were processed in a 3:1 mixture of H2SO4/HNO3 for 12h at ambient temperature and washed with deionized water. Subsequently, a ZnO thin film (30nm) was deposited on CNT fiber by atomic layer deposition (ALD) to serve as seed layer. One advantage of ALD technology is the deposited materials are chemisorbed on the surface of various substrates, therefore the excellent interface quality between seed layer and CNT fiber guarantee that the ZnO NWs could uniformly grow on the surface of CNT fiber. In the process of ALD, diethylzinc [Zn(CH<sub>2</sub>CH<sub>3</sub>)<sub>2</sub>] (denoted DEZn henceforth provided by J&K) and deionized water, which were used as metal and oxidant precursors, were introduced into the growth chamber separately. The process temperature and pressure were 150°C and 0.2 torr, respectively. Ar gas was used as carrier and purging gas to remove the residues and by-products. The deposition is achieved by repeating the following pulse sequence: t1-t2-t3-t4-t5-t6 up to the desired thickness, where "t1-t2-t3-t4-t5-t6" are the individual pulse times of DEZn, reaction, Ar, H<sub>2</sub>O, reaction and Ar, respectively. All the pulse times are given in seconds. During the ZnO ALD on CNT fiber, the timing sequence are 0.02-5-30-0.02-5-30. After 120 ALD cycles, a ZnO seed layer ( $\sim$ 30nm) were deposited on the surface of the CNT fiber.

### 2. Figures



Figure S1. X-ray diffraction pattern of CNT@ZnO@MnO<sub>2</sub> hybrid fibers.



Figure S2. Ragone plot of the FASCs devices:  $E_{V}$  and  $P_{V}$  based on the total volume and compares performance with that for other fiber-shaped energy storage devices.