Supporting Information

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The TEM image in Fig. S1a reveals the morphology of a catalyst nanoparticle, locating at the tip of an as-grown CNF. The metal catalyst nanoparticle used in the growth of CNFs possesses a polyhedral shape, which may explain the formation of polygonal CNFs. The corresponding TEM-EDX spectrum (see Fig. S1b) shows Ni, Sn, Cu, C, and O peaks, in which Ni and Sn signals originate from the catalyst particles, while C and Cu signals are attributed to the carbon-coated copper grids used in TEM characterization, further confirming the composition of catalysts used in PCNF growth.

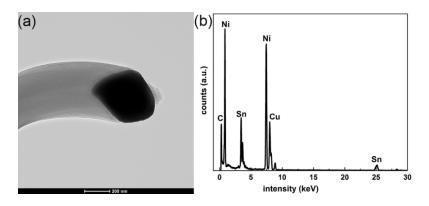


Fig. S1 Catalyst on the tip of a fiber: (a) TEM image; (b) EDX spectrum.

Fig. S2a displays optical images of carbon products obtained after 60 minutes of CVD growth. The left one shows a few catalyst particles (the white particles) in the boat. However, after 60 minutes of growth, the boat was fully filled with black carbon products. The relationship between carbon yield and growth time is revealed in Fig. S2b. The relative carbon yield (the weight ratio of carbon products to catalysts) increases steadily as the growth time prolongs. Even after a long-time growth, the catalyst still remains its catalytic activity, leading to a high carbon yield.

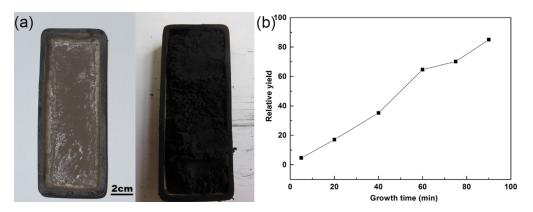


Fig. S2: (a) the optical images of the reactive boat covered with catalysts before (the left one) and after 60 minutes of CVD process (the right one); (b) relative carbon yield vs. growth time.

Fig. S3a displays a crack on the surface of a raw PCNF. Some edges connected weakly, forming narrow crevices on the surface. After activation by KOH at a KOH:C weight ratio of 4:1, some PCNFs were partially etched, and cracks or broken ends were clearly observed (see Fig. S3b). The inset in Fig. S3b shows that the carbon atoms were arranged sparsely in the core. Moreover, the fiber walls of PCNFs became thin after KOH etching, as shown in Fig. S3c.

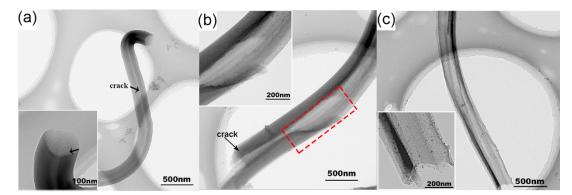


Fig. S3 TEM images of (a) raw PCNFs; (b) and (c) a-PCNFs-4 (the red rectangular box indicates the core structure of PCNFs).

Fig. S4 shows the galvanostatic charge-discharge curves of different PCNF-based electrodes at a current density of 3A g⁻¹ with a potential range of 0~1 V vs. SCE. The linear charge-discharge curve of raw PCNFs further confirms the double-layer capacitance. Nevertheless, voltage plateaus appeared in the discharge curves of the samples after activation, matching well with the peaks observed from the CV curves, which further proves the pseudocapacitive behavior of the a-PCNF electrodes. The specific capacitances of the raw PCNF, a-PCNFs-2, a-PCNFs-3 and a-PCNFs-4 electrodes were calculated to be 27.3, 103, 126, and 187 F g⁻¹ from the galvanostatic charge-discharge curves, respectively.

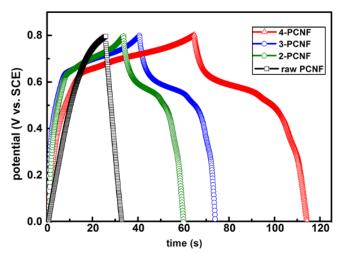


Fig. S4 galvanostatic charge and discharge curves of different PCNFs-based electrodes measured with a current density of 3 A g⁻¹.

Fig. S5 displays the peak current vs. scan rates of a-PCNFs-4 electrodes. The current response to an applied sweep rate reflects whether the redox reaction is diffusion-controlled or surface-controlled (capacitive process).¹ For a redox reaction limited by semi-infinite linear diffusion, the peak current varies with scan rate^{1/2}, but for a capacitive process, the peak current varies directly with scan rate. From this plot, the peak currents of both anode and cathode are directly proportional to the scan rate, indicating the redox reactions that take place in a-PCNFs-4 electrodes are surface-controlled.

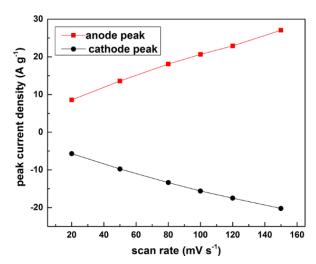


Fig. S5 the relationship between peak current and scan rate.

References

1 P. Simon, Y. Gogotsi and B. Dunn, *Science Magazine*, 2014, 343, pp. 1210-1211.