

Electronic Supplementary Information

## **Microwave plasma-induced graphene-sheet fibers from waste coffee grounds**

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### **Experimental section**

#### **Synthesis of Graphene-Sheet Fibers**

The waste coffee grounds (The remainder of regular coffee powder after being boiled, UCC) was dried at 100 °C for 6 hours without other treatments. Graphene-sheet fibers (GSFs) were fabricated from dried coffee grounds loaded in a nickel case using 2.45 GHz microwave plasma system at the power of 900 W, which was equipped with a rectangular waveguide to couple the microwave through quartz tube for generating the plasma. In the system, no additional heater was installed for substrate heating. The

substrate temperature was controlled by microwave power and plasma exposure time, and was measured by a thermocouple placed in direct contact with the substrate holder. After the deposition chamber was pumped down to a base pressure of 1 Torr by a rotary pump with the pumping gas rate of 300 L/s, the hydrogen and argon gases were introduced into the chamber and produced the plasma to irradiate the coffee grounds for 15 min. The maximal temperature was 650 °C during the process of microwave plasma irradiation. After deposition, the plasma was shut down and the equipment was naturally cooled to room temperature. The yield of GSFs was found to be 10-20% in all produced nanocarbons.

### **Characterization**

Morphologies and microstructures were investigated using a Hitachi SU8000/8020 scanning electron microscopy at 15 kV, a JEM2100 aberration-corrected transmission electron microscopy at 80 keV, a JEM2100F transmission electron microscopy at 200 kV, and an inVia Renishaw Raman spectroscopy with a laser wavelength of 532 nm. Chemical composition was performed by electron dispersive X-ray analyzer (EX-64175JNU)

### **Electrical Measurement**

The whole current-voltage measurements were carried out in the scanning electron microscope (JSM 5600) chamber with a high vacuum of  $\sim 10^{-6}$  Torr at room temperature. Two PtIr needles were employed as probes.

### **Electrochemical Measurement**

Cyclic voltammetric measurements were carried out at a CHI 600C electrochemical workstation assembled with a Pt wire as counter electrode, an Ag/AgCl (3 M KCl) electrode as reference electrode, and a glassy carbon (GC,  $\text{\O} = 3$  mm), Pt ( $\text{\O} = 1.6$  mm) or graphene-modified Pt electrode as working electrode. Prior to modification, the GSFs were collected slightly by the micro spatula and dispersed ultrasonically in the solution of dimethyl formamide. And the Pt electrode ( $\text{\O} = 1.6$  mm) was polished with 0.05  $\mu\text{m}$  alumina slurry and then washed by deionized water. Sequentially, the clean Pt with some ethanol was directly covered by graphene film in a rectangle shape (2 mm $\times$ 3 mm), which was modified by a cut. Finally, the graphene-modified Pt was dried by a halogen lamp for 30 min. All potentials were reported in this context with respect to the reference electrode of Ag/AgCl (3 M KCl). Before the electrochemical experiments, all solution to be measured was purged with nitrogen gas for about 10 min.

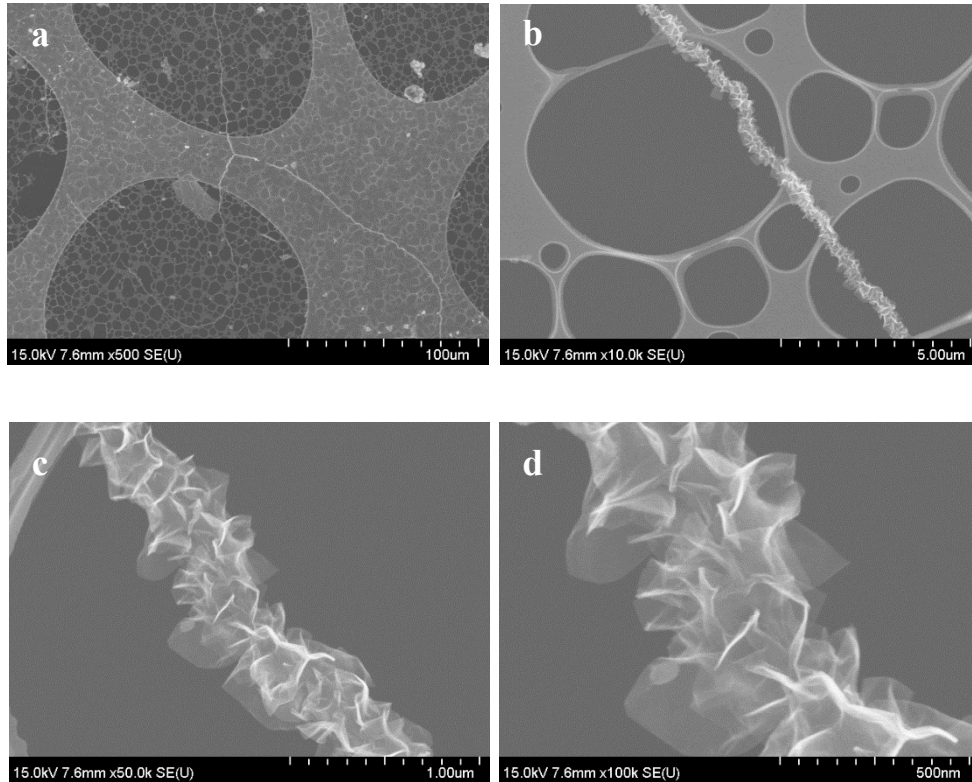


Figure S1 Morphologies of the GSFs transferred on the Cu grid with different magnifications. (a)  $\times 500$ , (b)  $\times 10\text{k}$ , (c)  $\times 50\text{k}$ , and (d)  $\times 100\text{k}$ .

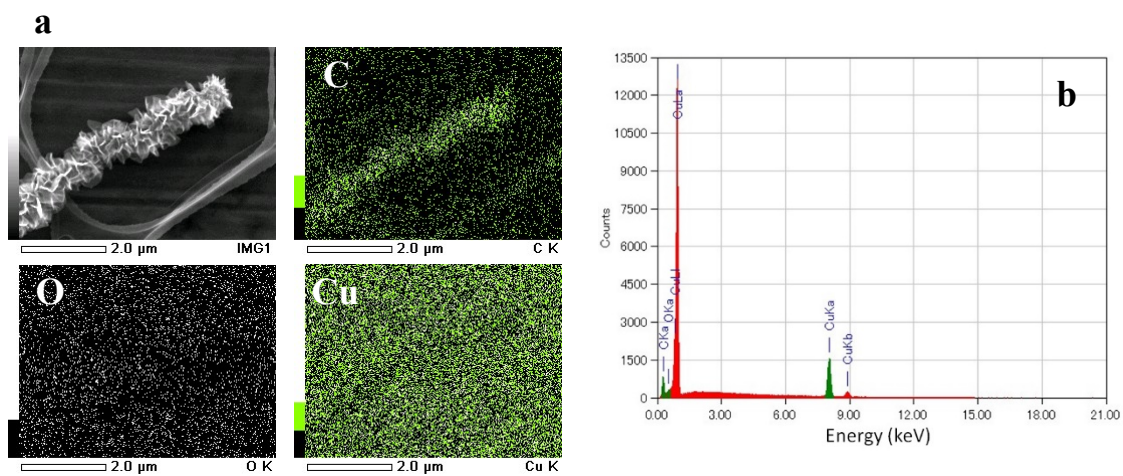


Figure S2 Chemical composition analyses of top region of individual GSF. (a) Energy dispersive x-ray mapping. (b) Corresponding energy dispersive x-ray spectrum. Both copper signals ascribe to the copper mesh grid.

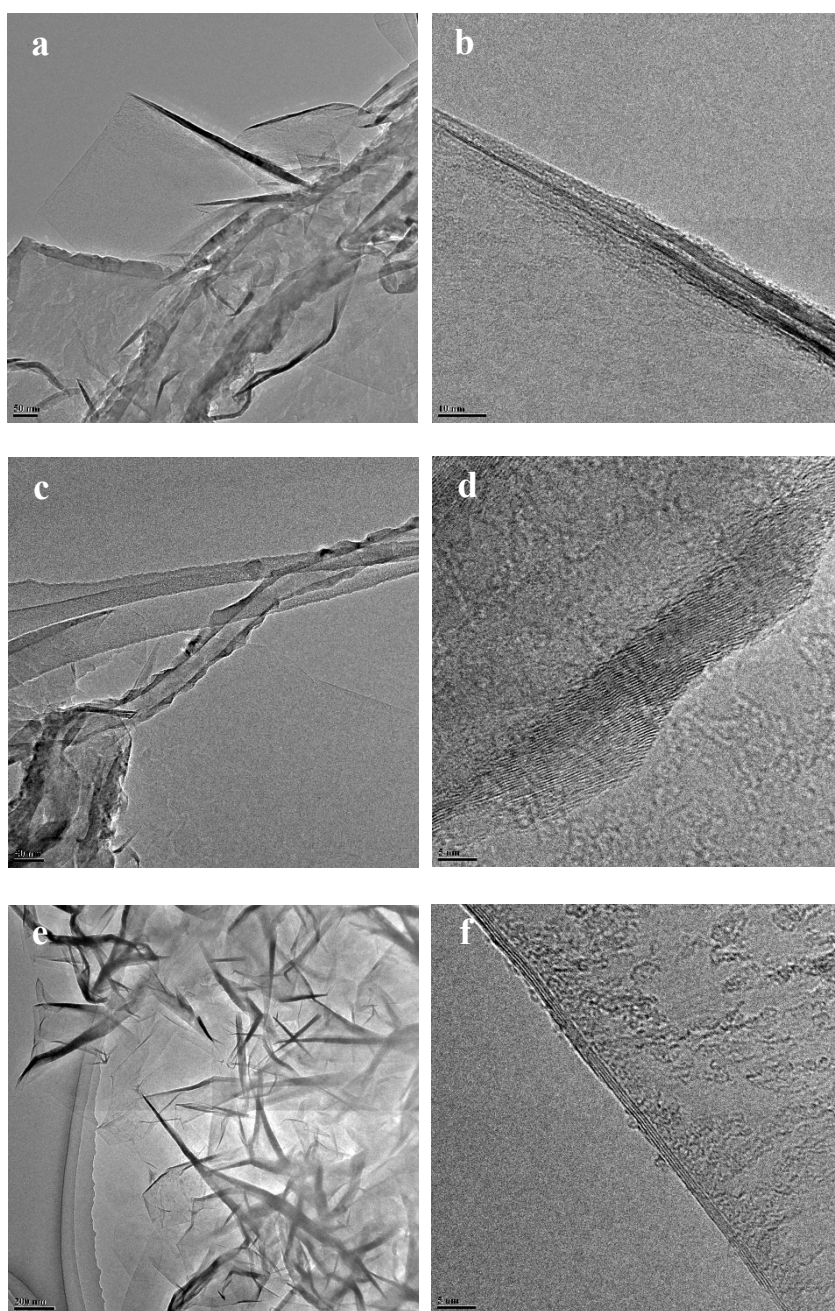


Figure S3 Microstructures of coffee-ground-derived nanocarbons by MPI technique. (a)-(b) graphenated carbon nanotubes consist of hollow structures, in which the sidewalls were decorated by few-layer graphene. (c)-(d) common carbon nanotubes show herringbone-like structures. (e)-(f) Particle-like carbon is composed of few-layer graphene.