

## Supporting Information

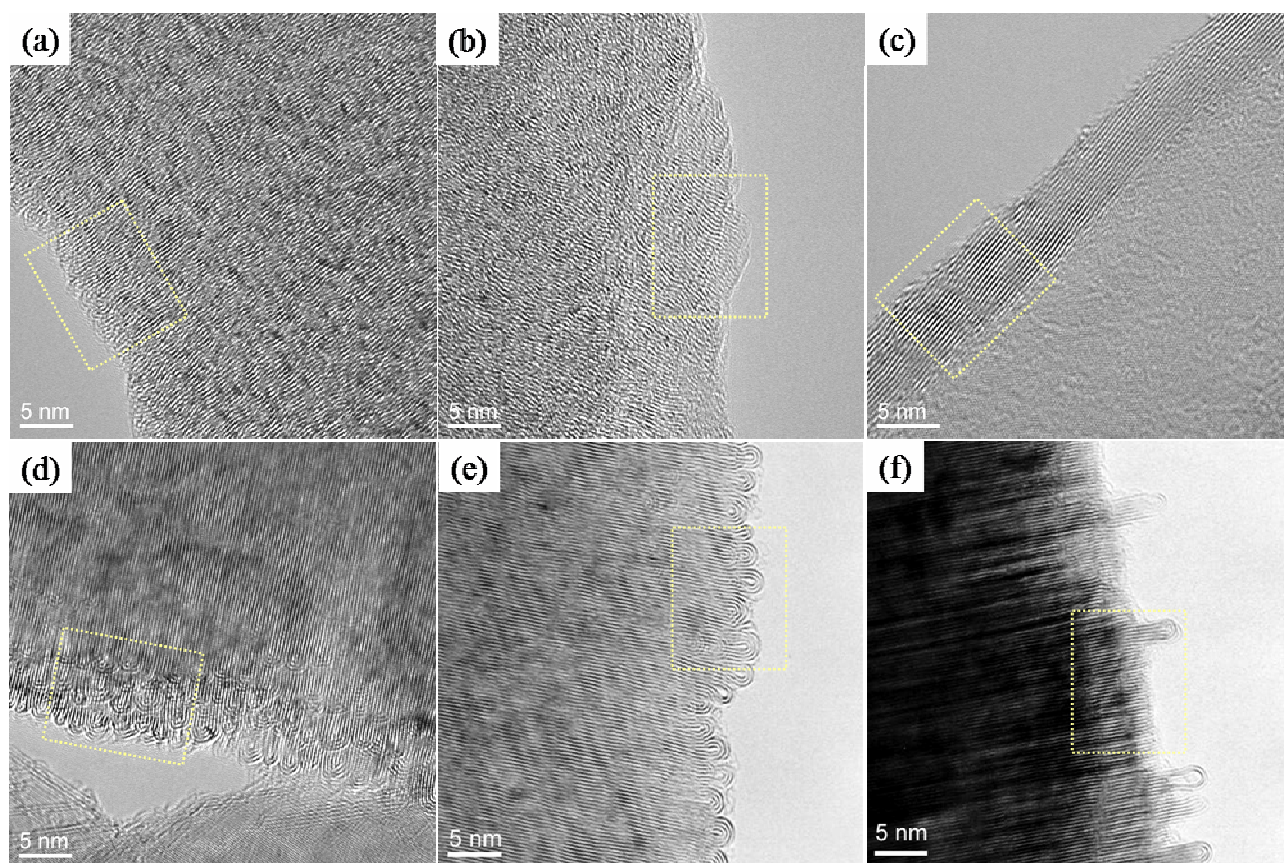
### Electrochemical Capacitances of Well-defined Carbon Surfaces

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#### ● Preparation of well-defined carbon nanofibers (CNFs) and modified platelet-type CNFs

The followings are the specific methods to prepare the carbon nanofibers (CNF) used in this study. The pristine CNFs, i.e. platelet-type CNF (PCNF), herringbone-type CNF (HCNF) and tubular-type CNF (TCNF) were synthesized from a CO/H<sub>2</sub> mixture or a C<sub>2</sub>H<sub>4</sub>/H<sub>2</sub> mixture on the catalyst of Fe, Ni-Cu (8:2), or Fe-Ni (2:8).<sup>S1-S3</sup> GPCNF was obtained by graphitization of pristine PCNF at 2800 °C for 10 min under an Ar atmosphere (GPCNF). The GPCNF was modified further by mechanically milling in ethanol for 72 h with stainless steel balls of 2 mm in diameter to produce GPCNF-M. To prepare GPCNF-NA, on the other hand, GPCNF was oxidized for 24 h with a 10% HNO<sub>3</sub> solution at room temperature. Details of the GPCNFs were fully documented elsewhere.<sup>S4</sup> The squares in S-Figure 1, accompanied with the corresponding schematic cartoons, are shown in Figure 1 of the main text.



S-Figure 1. TEM images of the CNFs: (a) PCNF, (b) HCNF, (c) TCNF, (d) GPCNF, (e) GPCNF-M, and (f) GPCNF-NA.

### ● Electrochemical measurement of capacitance

For capacitance measurements, appropriate amounts of CNFs, water and 5% Nafion solution (Wako, Japan) were mixed and sonicated to produce CNF slurries. A certain aliquot of the slurry was spread uniformly on a Au disk, and the uniform layer was dried under an IR lamp for 20 min. After immersing the layer of CNF slurry on the Au electrode into 0.5 M H<sub>2</sub>SO<sub>4</sub> solution (Johnson Matthey, ACS grade, USA), a conventional three electrode system, using a Ag/AgCl electrode ([Cl<sup>-</sup>] = 1 M) as a reference electrode and a Pt gauze (Johnson Matthey, 52 mesh, 25 × 25 mm<sup>2</sup>, USA) as a counter electrode, was employed to obtain a cyclic voltammogram at the scan rate of 10 mV/sec. By measuring the capacitive current at 0.45 V, the capacitance of a CNF was estimated. The amount of the slurry, transferred onto the Au disk electrode, was constant within 5% error, and the weight of the CNF was estimated from the volume of the slurry to normalize the capacitance value against weight. The potential values in the main text are the one as measured.

### References for Supporting information

- (S1) Tanaka, A., Yoon, S. H., Mochida, I. *Carbon* **2004**, 42 (3), 591-597.
- (S2) Tanaka, A., Yoon, S. H., Mochida, I. *Carbon* **2004**, 42 (7), 1291-1298.
- (S3) Yoon, S. H., Lim, S., Song, Y., Ota, Y., Qiao, W., Tanaka, A., Mochida, I. *Carbon* **2004**, 42 (8-9), 1723-1729.
- (S4) Lim, S., Yoon, S. H., Mochida, I., Chi, J. H. *J. Phys. Chem. B* **2004**, 108 (5), 1533-1536.