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Supplementary Material

## Three-dimensional porous polyaniline/graphene-coated activated carbon fiber electrodes for supercapacitors

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1. Supplement to microstructures and surface morphologies of Ni-coated ACFs, FLG/Ni-coated



## ACFs, and PANI/FLG-coated ACFs

Fig. S1. FESEM images of Ni-coated ACFs at various electrodeposition times: (a) 5 min, (b) 10 min,

(c) 15 min, and (d) 45 min.



Fig. S2. FESEM images of FLG-coated ACFs at various electrodeposition times: (a) 10 min, (b) 15 min, and (c) 45 min.



Fig. S3. (a) AFM image of FLG from FLG-coated ACFs-30; (b) SEM image of FLG-coated ACFs-30; TEM images of (c) FLG peeled off from FLG-coated ACFs-30 and (d) PANI/FLG peeled off from PANI/FLG-coated ACFs-30.



Fig. S4. (a) FTIR and (b) Raman spectra of desized ACFs, FLG/Ni coated ACFs, and PANI/FLG coated ACFs.

Fig. S4 depicts the FTIR spectra of desized ACFs, FLG/Ni-coated ACFs, and PANI/FLGcoated ACFs. The spectrum of the desized ACFs shows intense bands at 3430 cm<sup>-1</sup> (corresponding to hydroxyl groups), 1625 cm<sup>-1</sup> (for C=C stretching), and 1383 cm<sup>-1</sup> (for C–H3 deformation). Owing to the relatively small amount of FLG and Ni particles in FLG/Ni-coated ACFs, the spectrum of FLG/Nicoated ACFs exhibits the similar curve to the desized ACFs. The additional peaks were observed in spectrum of PANI/FLG-coated ACFs, corresponding to C-C-N bending (around 1191 cm<sup>-1</sup>), C-N stretching (around 1165 cm<sup>-1</sup>), quinoid ring stretching (around 1503 cm<sup>-1</sup>), and benzoid ring stretching (shoulder around 1584 cm<sup>-1</sup>).

The structural of the desized ACFs, FLG/Ni-coated ACFs, and PANI/FLG-coated ACFs were studied by Raman spectra (Fig. S4b). As compared with the spectrum for FLG/Ni-coated ACFs, the higher  $I_D/I_G$  ratio of 0.73 for the desized ACFs indicated the presence of more defective amorphous carbon structures. Owing to the PANI coverage on PANI/FLG-coated ACFs, only PANI spectrum was obtained. The characteristic peaks of PANI were observed, corresponding to quinone (around 1211, 1473, and 1490 cm<sup>-1</sup>), C-N in quinoid ring (around 1211 cm<sup>-1</sup>), C-N and C-C in benzoid ring (around 1345 cm<sup>-1</sup>), C=C in both quinoid and benzoid rings (around 1591 cm<sup>-1</sup>). 2. Supplement to electrochemical properties of ACFs and PANI/FLG-coated ACFs



Fig. S5. CV comparison of ACFs, PANI film, and FLG film electrode at a scan rate of 5 mV s<sup>-1</sup>.



Fig. S6. IR drop in charge–discharge curves of (a) ACFs and (b) PANI/FLG-coated ACFs-30 at a charging current of  $0.1 \text{ A g}^{-1}$ .