Electronic Supplementary Information

**Heterogeneous mesoporous NiCo\(_2\)O\(_4\)-MnO\(_2\)/graphene foam asymmetric supercapacitors with ultrahigh specific energies**

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Fig. S1 (a) Digital photograph, (b-d) SEM images of cellular-structured freestanding GF network, (e, f) TEM analysis of GF, (g, h) CNTs grown on GF substrate (CNT/GF) by CVD as the negative electrode in asymmetric supercapacitors, and (i) typical Raman spectra of GF and CNT/GF hybrid.
Fig. S2. EDS line scan analysis of NiCo$_2$O$_4$-MnO$_2$ core-shell structure.
Fig. S3 (a) nitrogen adsorption/desorption isotherm curve and (b) pore size distribution of NiCo$_2$O$_4$/GF.

Fig. S4 Nitrogen adsorption/desorption isotherm curve of GF.
Fig. S5a shows the general XPS spectra of NiCo$_2$O$_4$-MnO$_2$/GF composite. The XPS spectrum for Mn 2p (Fig. S5b) has two well-defined peaks separated by ~11.5 eV. They are assigned to the spin-orbit doublets of Mn 2p1/2 (at 653.5 eV) and Mn 2p3/2 (at 641.8 eV). They are characteristic of Mn(IV) oxidation state, consistent with the reduction reaction of KMnO$_4$ mentioned above. The deconvoluted spectra of Co 2p in Fig. S5c also shows two peaks at 779.8 and 794.7 eV assigned to Co$^{2+}$ along with two other peaks at 780.6 and 795.8 eV corresponding to Co$^{3+}$ as well as two flat satellites at 785.6 and 804 eV. Fig. S5d shows the Ni 2p spectra with two Ni$^{2+}$ spin-orbit peaks at 854.3 and 871.6 eV and another doublet of Ni$^{3+}$ at 855.4 and 872.8 eV. Each doublet was accompanied by a shake-up satellite at 861.2 and 879.5 eV binding energies. The deconvoluted C 1s spectra (Fig. S5e) consisted of a sharp peak at 284.4 assigned to the C-C bonds and a small peak at 285.3 eV corresponding to the C-H bonds. The deconvoluted O 1s spectra (Fig. S5f) presented a sharp peak located at 529.7 eV attributed to the O-M bonds, while the shoulder at 531.7 eV is attributed to the surface oxygen defects or adsorbed water on the surface.
Fig. S5 (a) The general XPS spectra and deconvoluted high resolution spectra of (b) Mn 2p, (c) Co 2p, (d) Ni 2p, (e) C 1s and (f) O 1s for NiCo$_2$O$_4$-MnO$_2$/GF composite.
Fig. S6 (a) CV curves measured at different scan rates, (b) GCD results and (c) gravimetric and areal capacitances measured at different current densities for the CNT/GF electrode.

