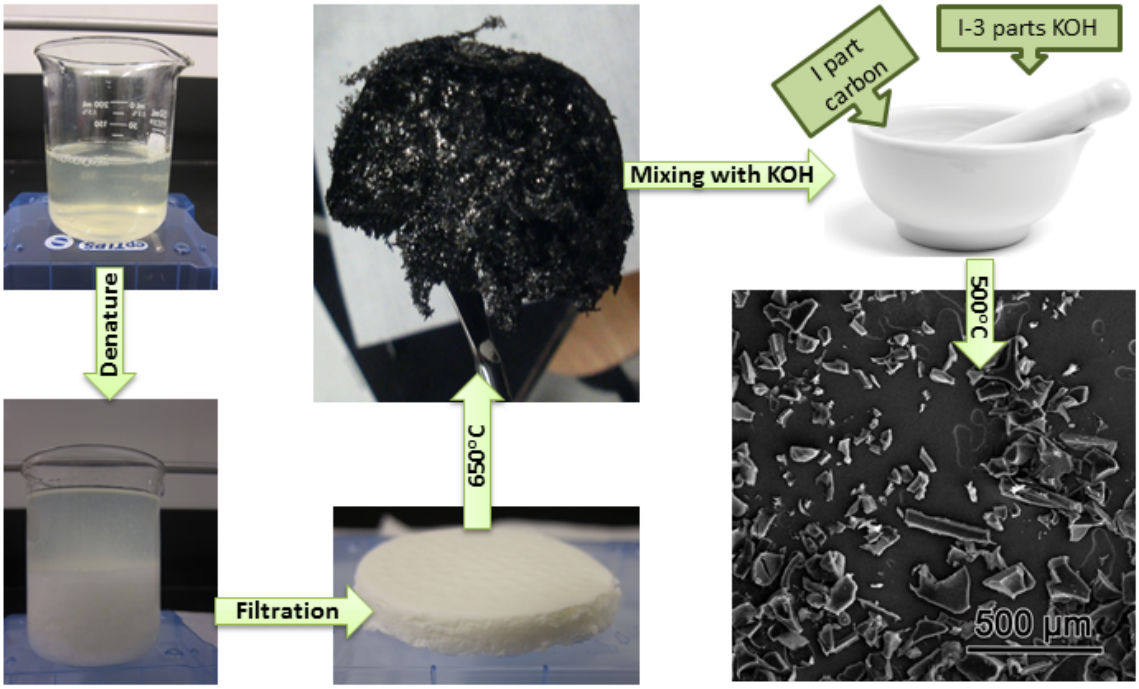
Electronic Supplementary Material (ESI) for Energy & Environmental Science.



Scheme S1. Preparation process for highly functionalized activated carbons (HFAC).

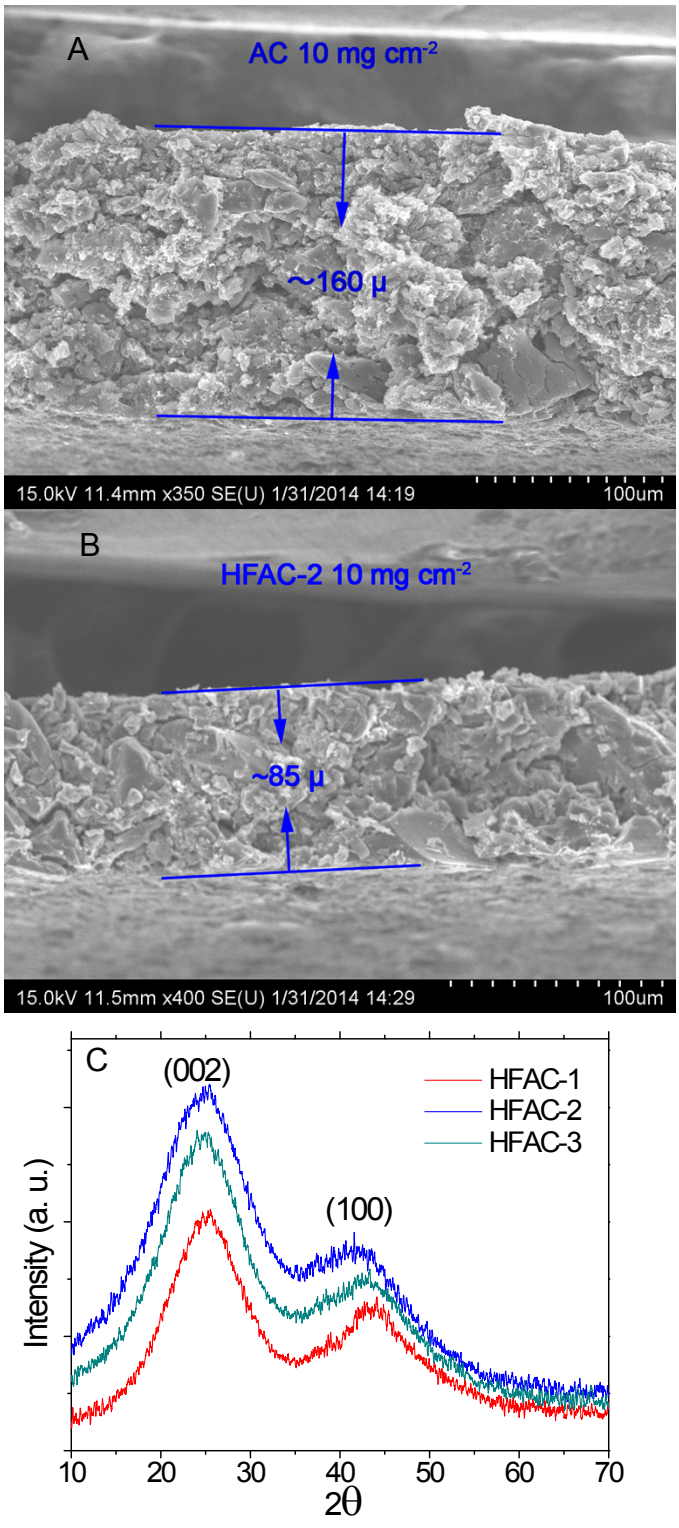


Figure S1. SEM images of AC (A) and HFAC-2 (B) electrode pressed at 100 MP; (C) X-Ray diffraction patterns of HFAC carbons.

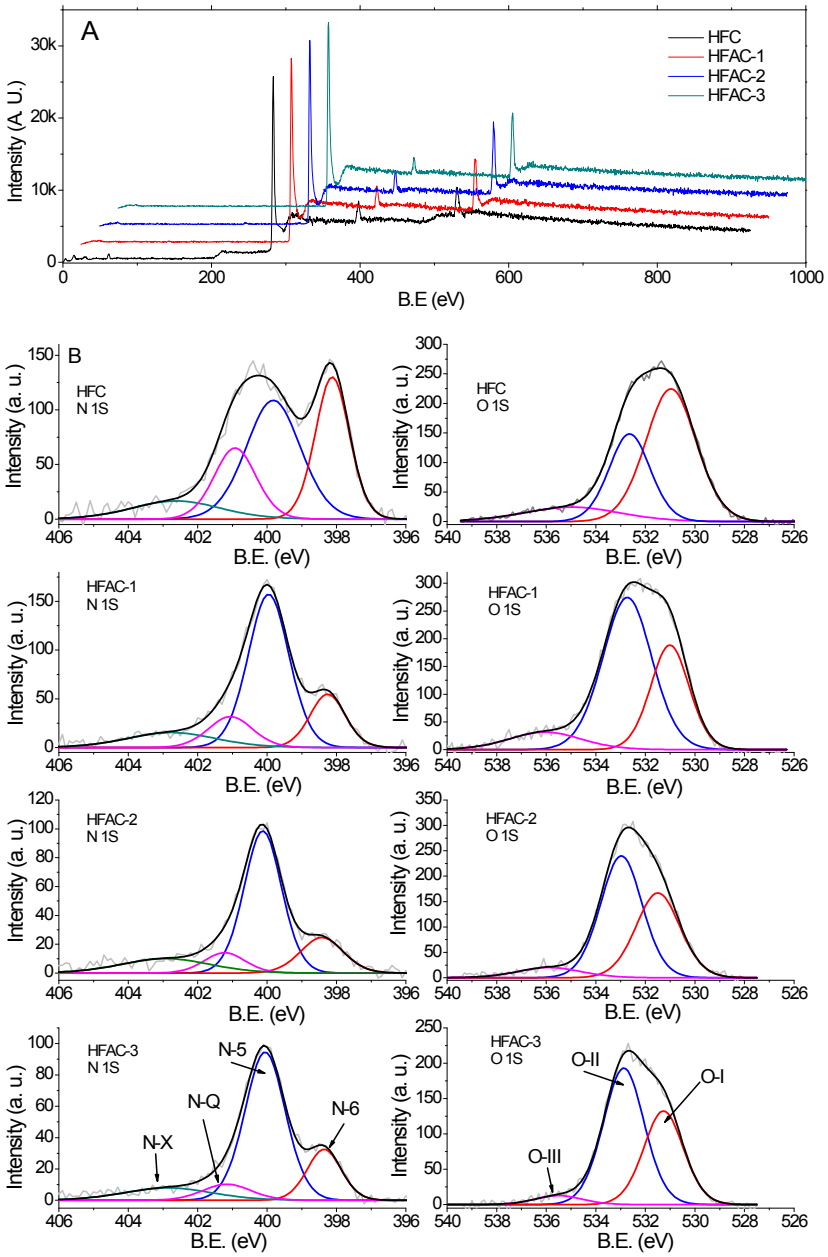


Figure S2. XPS spectra of HFAC materials. (A) XPS survey from 0 to 1000 eV; (B) N 1S and O 1S core level XPS spectra fitting.

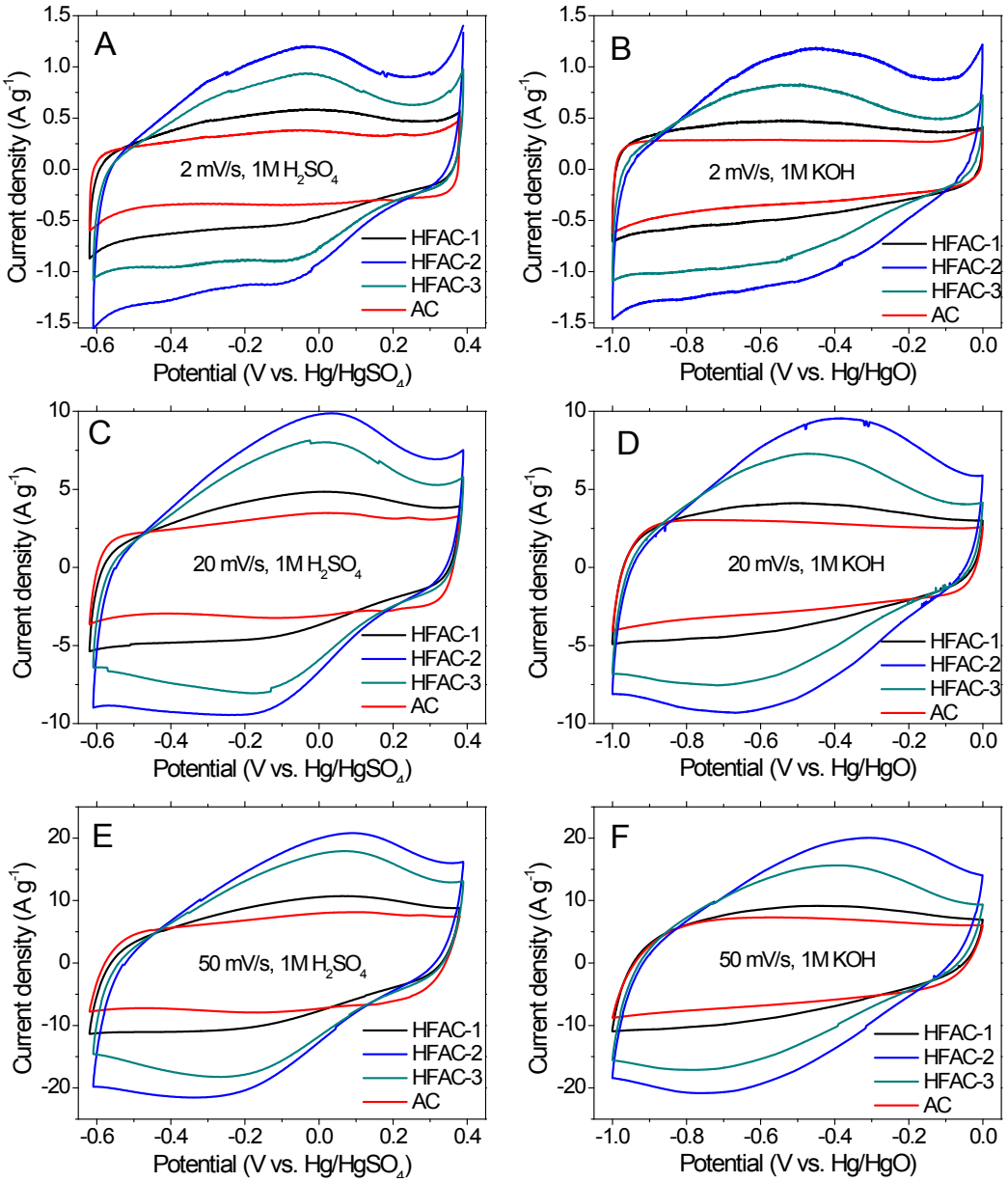


Figure S3. Cyclic voltammograms (CVs) of HFAC and of the baseline AC in 1M H_2SO_4 (A, C, E) and in 1M KOH (B, D, F) at 2, 20 and 50 mV s^{-1}.

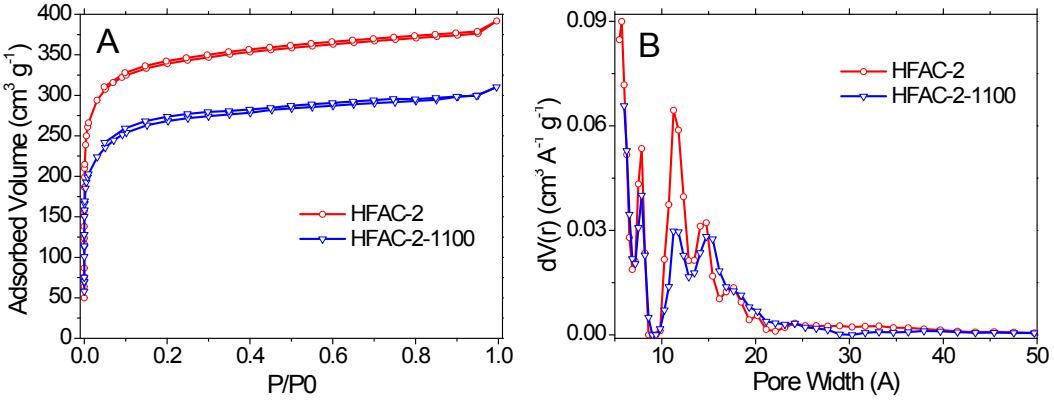


Figure S4. (A) Nitrogen adsorption isotherms at 77k and (B) NLDFT pore-size distribution of HFAC-2 before and after an 1100°C anneal in Ar to eliminate the functional groups.

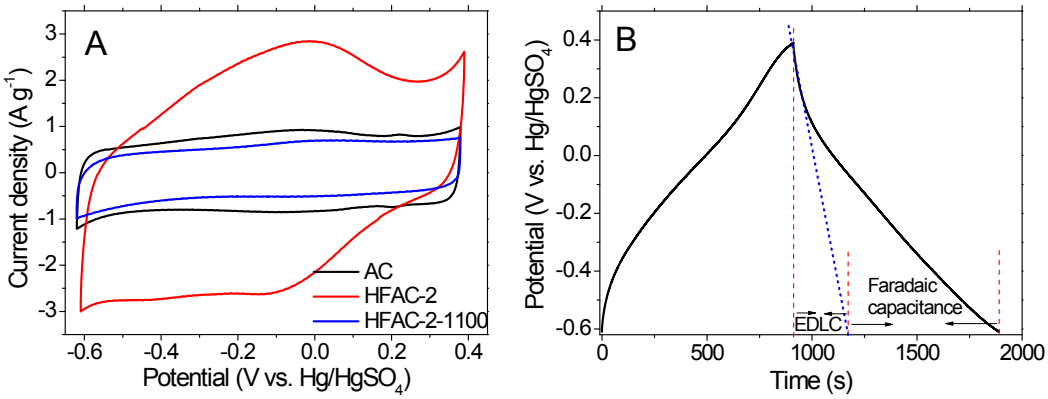


Figure S5. (A) Cyclic voltammograms (CVs) of HFAC-2 before and after an 1100°C anneal in Ar to eliminate the functional groups, AC being included as a baseline; (B) galvanostatic charge/discharge curve of HFAC-2 at 0.5 A g^{-1} with the estimated EDLC and faradaic capacitance contributions being obtained from the discharge portions of differing slope. Testing was done in 1M H_2SO_4 at 5mV/S.

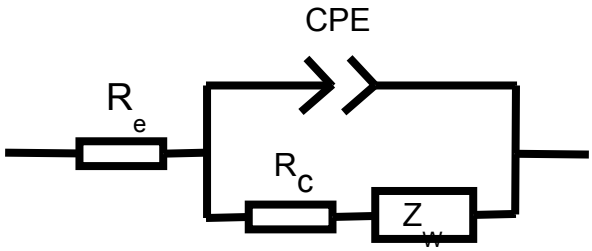


Figure S6. Corresponding equivalent circuit used for Nyquist plot simulation, where Re is the total resistance of electrolyte, electrode, current collector and separator, Rct is the charge transfer resistance, Zw is the Warburg impedance related to the diffusion of ions into the bulk electrode, CPE stands for constant phase element related to the capacitive behavior of electrodes.

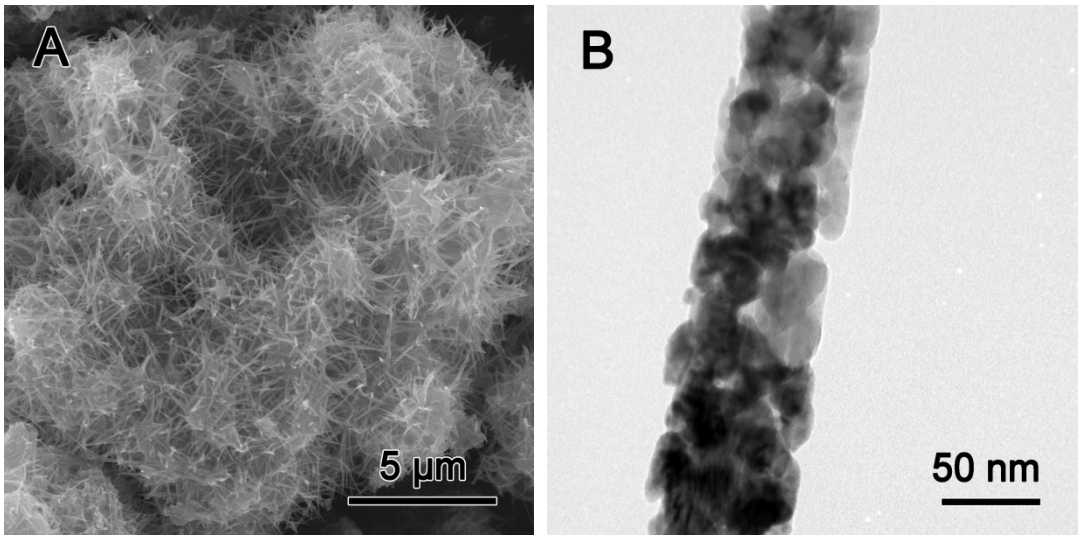


Figure S7. (A) SEM and (B) TEM micrographs of the $NiCo_2O_4$/graphene nanocomposite used as positive electrode in the asymmetric cell. Figure (B) shows a bright field image of a representative "nano-needle" of $NiCo_2O_4$ that is composed of 10-20 nm crystallites interspersed with mesoporosity.

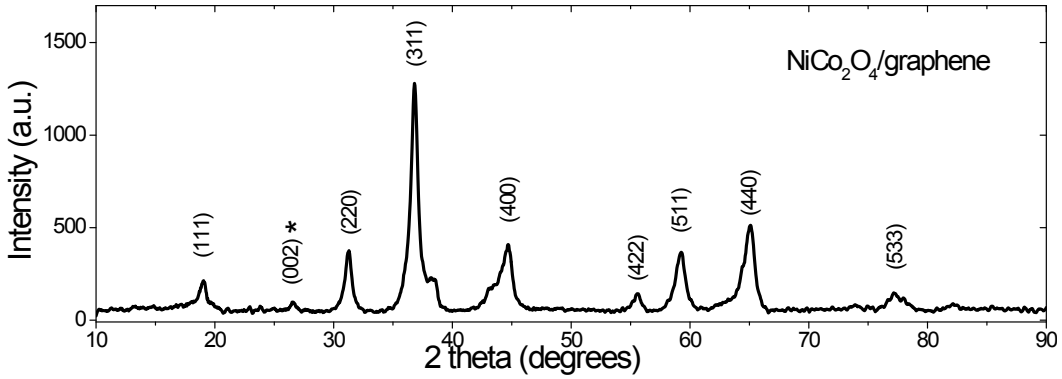


Figure S8. X-Ray diffraction pattern of $NiCo_2O_4$/graphene nanocomposite used as positive electrode in the asymmetric cell. The diffraction peak marked with * is indexed as (002) graphitic peak associated with the graphene nanoflakes, while the other peaks are indexed as equilibrium $NiCo_2O_4$.

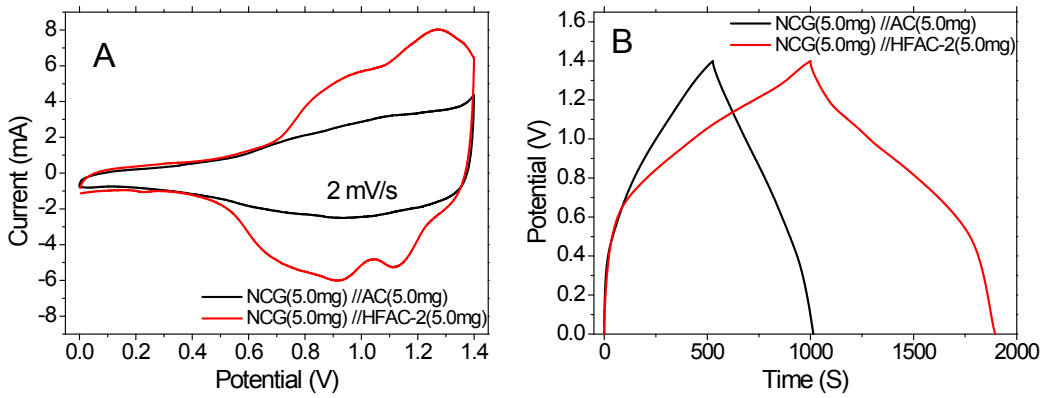


Figure S9. Comparative electrochemical performance of asymmetric supercapacitor cells, both of which employ 5 mg of the same positive electrode ($NiCo_2O_4$/graphene), but with a different negative electrode (5 mg HFAC-2 vs. 5 mg AC). (A) CVs of the two asymmetric full cells; (B) galvanostatic charge/discharge curves of the two asymmetric cells at 0.25A g^{-1}.