Supplementary Information

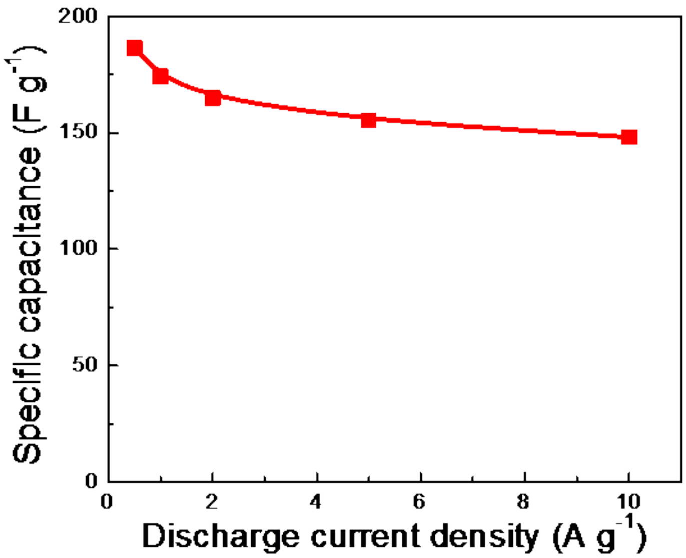

Figure S1.Specific gravimetric capacitance at different discharge densities of a-graphene electrode.

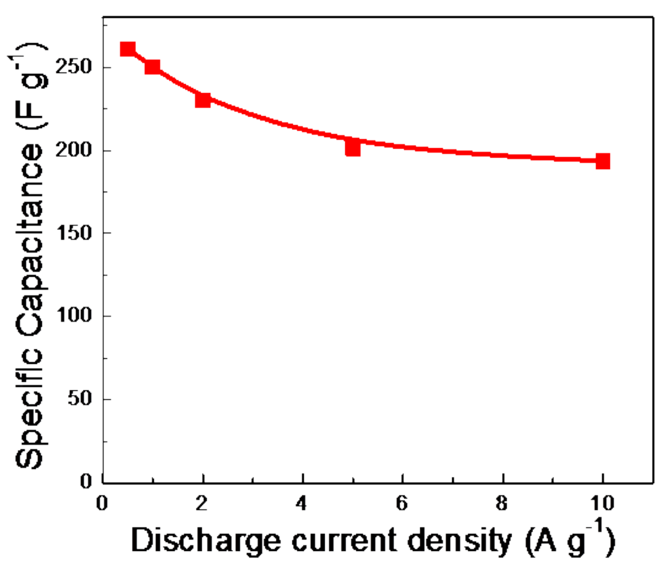

Figure S2. Specific gravimetric capacitance at different discharge densities of PEDOT/A-CNTs electrode.

In order to investigate the improvement of the A-CNTs on the cycle life retention of composite electrodes compared with randomly packed CNT networks, supercapacitors with random PEDOT/CNTs electrodes were also fabricated. The random CNTs electrodes were prepared by mixing 90wt% active materials and 10wt% polytetrafluoroethylene(PTFE) to form slurry.The slurry was pressed gold current collector, then dried at 80 ^{o}C for 24 h. 3,4-Ethylenedioxythiophene (EDOT) with molecular weight 142.17 g mol^{-1} was received from Sigma-Aldrich. Sodium poly(styrene sulfonate) (PSS) with molecular weight 70,000 was purchased from Sigma Aldrich. Electrochemical polymerization of PEDOT was performed using the Autolab PGSTAT 302N (Metrohm, USA) at current density 0.5 mA cm^{-2} in room temperature. PEDOT was grown on the electrodes and around the CNTs in an aqueous solution of EDOT (0.01 M in deionized water) and PSS (0.1 M in deionized eater) with a conventional four-electrode configuration. The amount of polymer coated on the electrode site was controlled by the total charge density passed during polymerization process (750 mC cm^{-2}, 1500 mC cm^{-2}, and 2250 mC cm^{-2}). The working and sensing electrodes were connected to the electrodes. The reference and counter electrode were connected to a platinum wire within the EDOT/PSS. The capacitance stability of PEDOT/A-CNTs and PEDOT/random CNTs symmetric supercapacitors was measured under driving voltages of 1 V and 2 V, respectively, at a current density of 5 A g^{-1} for 1,000 cycles as shown in Figure S3.

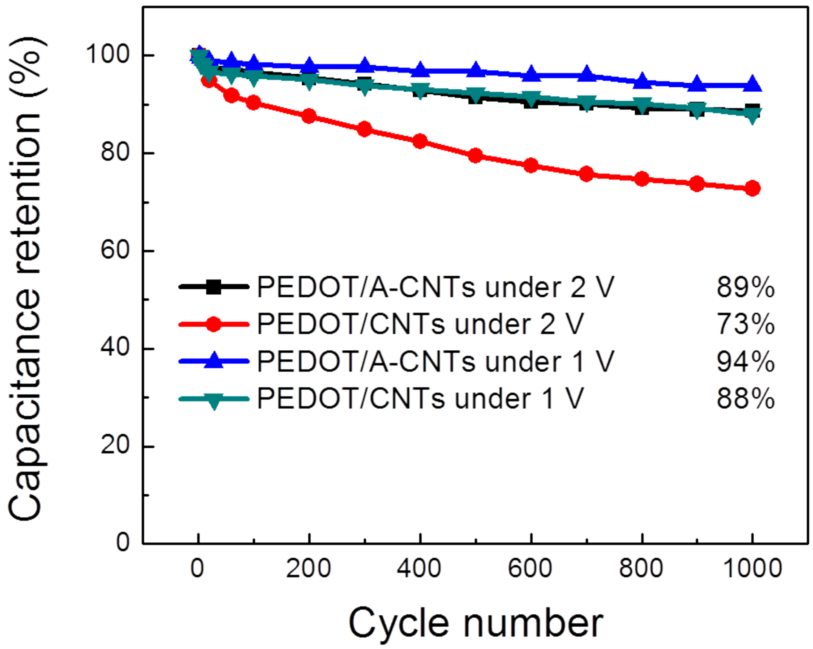

Figure S3. Cycle performances of symmetric supercapacitors with the PEDOT/A-CNTs electrodes and the PEDOT deposited on randomly packed CNT networks under driving voltages of 1 V and 2 V, respectively, at current density of 5 A g^{-1}.

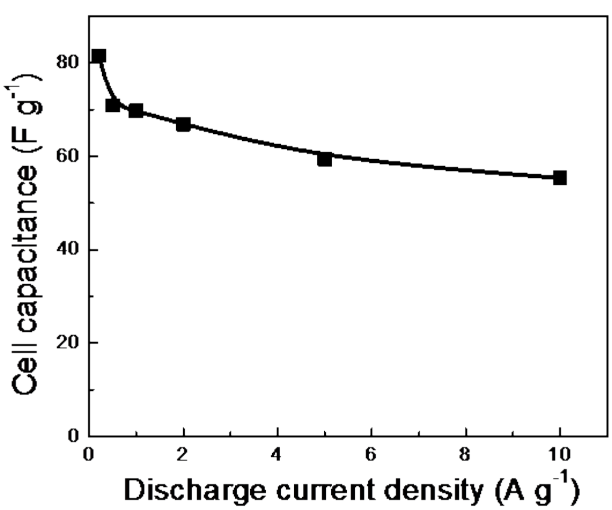

Figure S4. Cell gravimetric capacitance with different discharge current densities.

As a comparison to emphasize the importance of electrolyte on the operation voltage of the cell, the cell with organic electrolyte of 1 M Et_4NBF_4/PC was also assembled, because 1M Et_4NBF_4/PC is a widely used electrolyte for supercapacitor industry application. The electrochemical performance is shown in Figure S5 which shows an operating voltage window of 3V, much lower than that of IL based electrolyte of 2 M $BMIBF_4$/PC.

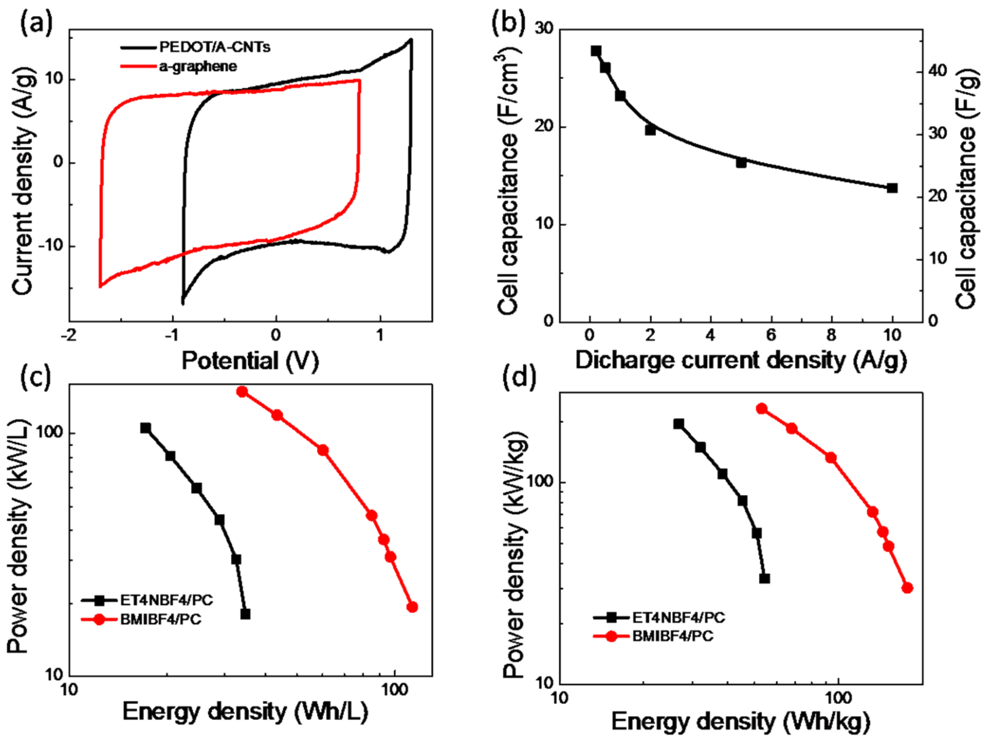

Figure S5. The electrochemical performance of cell using 1 M Et_4NBF_4/PC as electrolyte.

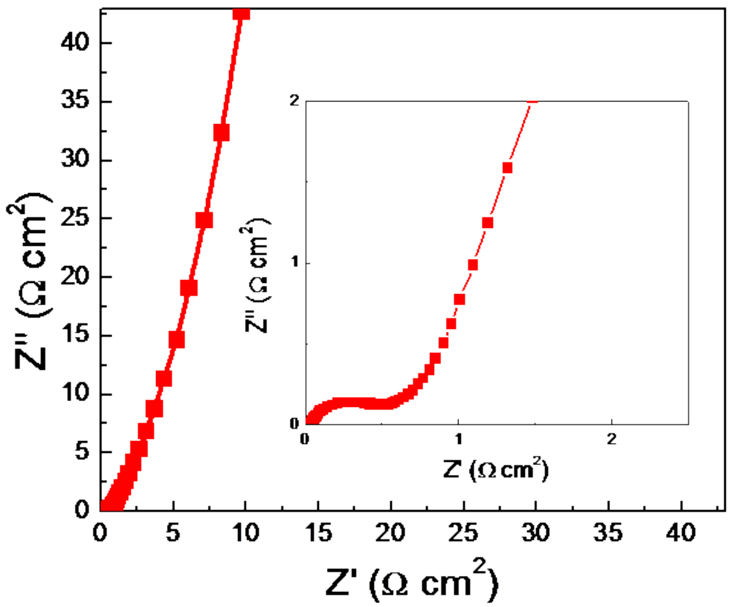

Figure S6.Nyquist plot of the asymmetric cell using 1 M $BMIBF_4$/PC as electrolyte.

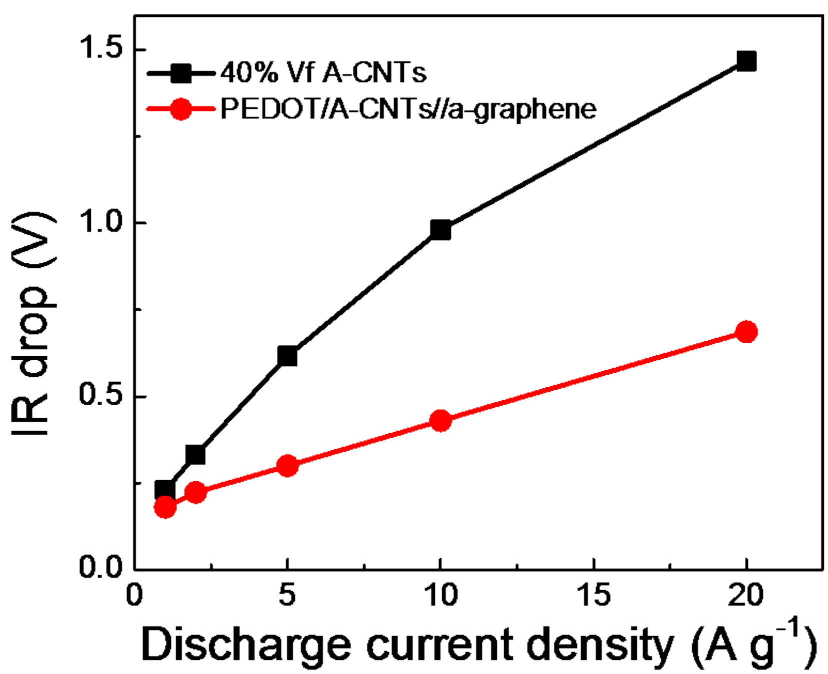

Figure S7. IR drop of cells with 40% Vf A-CNTs and PEDOT/A-CNTs//a-graphene as electrodes, respectively.