Supplementary Information

Non-polymeric hybridization of a TEMPO derivative with activated carbon for high-energy-density aqueous electrochemical capacitor electrodes

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Contents

Thermogravimetric analysis (TGA) of HTB.

For the determination of the sublimation temperature of HTB, TGA of HTB was performed with a thermogravimetric analyzer (DTG-60H, Shimadzu) under nitrogen flow. As shown in Figure S1, HTB sublimates at ca. 125 ºC.

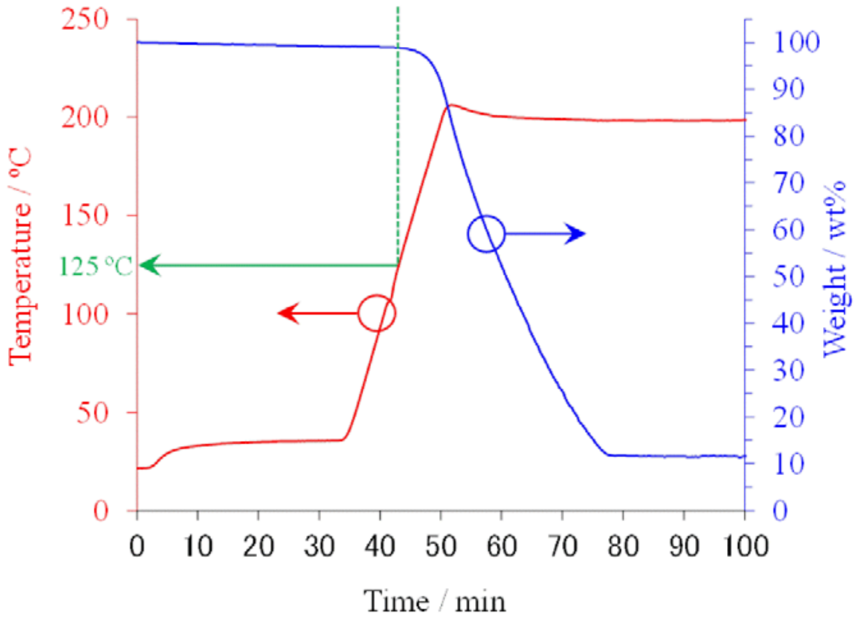

Figure S1. TGA pattern of HTB measured at a heating rate of 10 ºC min^{-1} under continuous nitrogen flow of 100 cc min^{-1}. The maximum temperature for the analysis was 200 ºC.

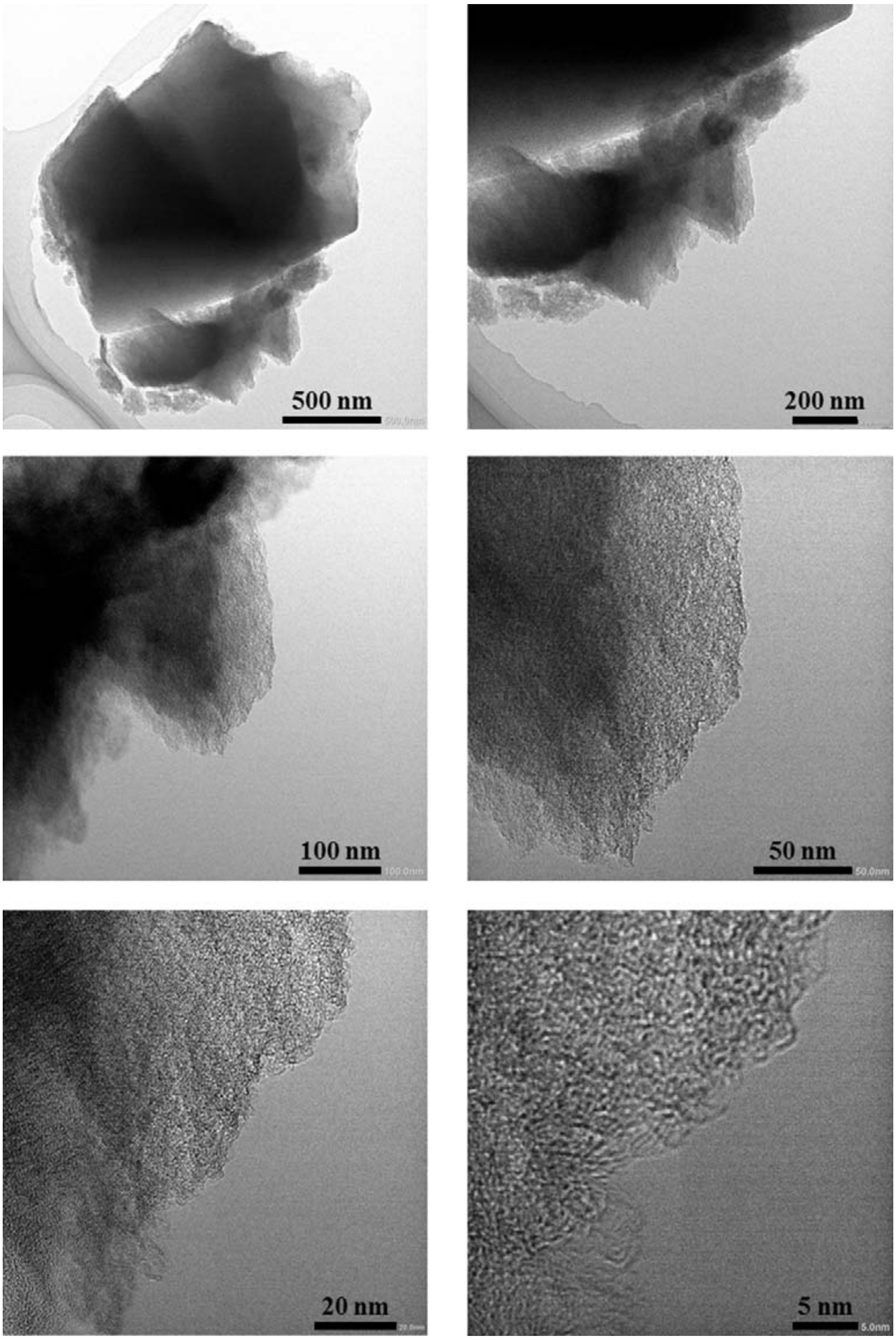

Figure S2. TEM images of AC/HTB (50%).

Cyclic voltammogram of HTB in 1 M H_2SO_4.

Since HTB is hydrophobic, 1 M H_2SO_4 was saturated with HTB and the resulting solution was analyzed by CV using a three-electrode cell. Pt wire was used as the working and counter electrode. A Ag/AgCl electrode (saturated KCl) was used as the reference electrode. The measurement was conducted at a sweep rate of 1 mV s^{-1} in the potential range from −0.1 to 0.9 V (*vs*. sat. Ag/AgCl) at 25 °C. For comparison, 1 M H_2SO_4 was also analyzed by CV. As shown in Figure S3, the voltammogram of HTB saturated in 1 M H_2SO_4 exhibits almost the same behavior as that obtained in 1 M H_2SO_4, and any redox peak was not observed, indicating that HTB does not undergo the redox reaction in 1 M H_2SO_4.

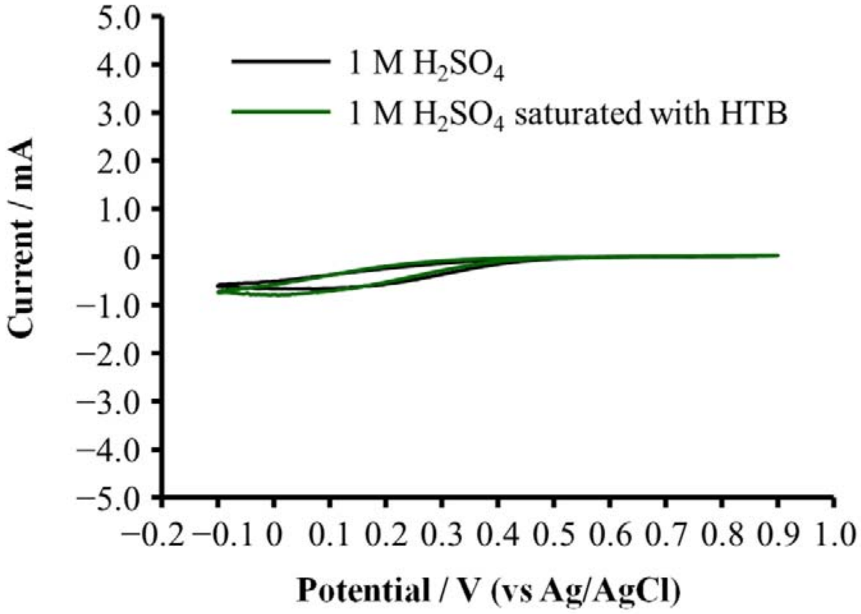

Figure S3. Cyclic voltammograms of 1 M H_2SO_4 saturated with HTB and 1 M H_2SO_4.

Cyclic voltammogram of HTB in 1 M Et_4NBF_4/PC.

For the examination of the electrochemical behavior of HTB in an organic electrolyte, the electrolyte with a concentration of 1 mM HTB and 1 M tetraethylammonium tetrafluoroborate (Et_4NBF_4) in propylene carbonate (PC) was prepared. Pt wire was used as the working and counter electrode. Ag/Ag^+ electrode using the acetonitrile solution with a concentration of 1 M Et_4NBF_4 and 0.1 M silver perchlorate ($AgClO_4$) was used as the reference electrode. The solution was analyzed by CV at a sweep rate of 1 mV s^{-1} in the potential range from 0 to 0.8 V (*vs*. Ag/Ag^+) at 25 °C. The voltammogram displays a distinct redox peak of HTB.

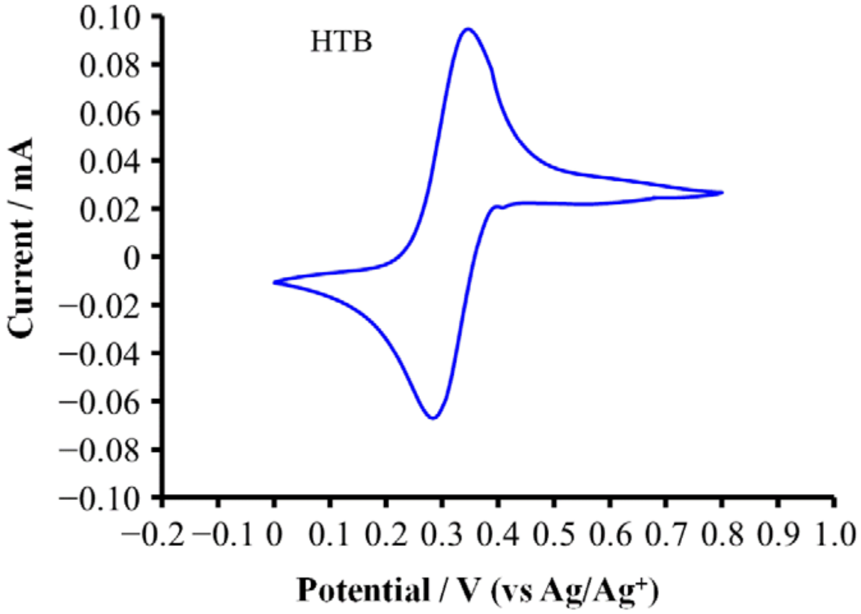

Figure S4. Cyclic voltammogram of HTB (1 mM) in 1 M Et_4NBF_4/PC.

Ragone plot of AC/HTB (30%)/AC hybrid asymmetric device.

For the measurement of the hybrid asymmetric device, the device was constructed from AC/HTB (30%) as the positive electrode and AC (MSC30) as the negative one with the same weight ratio. The constant power discharge measurements was performed in a cell voltage of 0.5 to 1.0 V at constant powers from 0.2 to 100 mW. The energy density was calculated from the constant power discharge curves from 1.0 to 0.5 V. As shown in the discharge profiles of AC/HTB (30%) and AC obtained from GC measurements at 1 A g^{-1} (Fig. 4c), their gravimetric capacities are almost the same, *ca*. 90 mAh g^{-1}. Therefore, the hybrid asymmetric device was prepared by using the same mass of each electrode. For comparison, the symmetric device consisting of AC was also evaluated. The hybrid asymmetric device exhibits a 1.3~1.5 time enhancement of the gravimetric energy density compared to the symmetric device (< 600 W kg^{-1}). Moreover, a 1.6~1.7 time enhancement was achieved for the hybrid asymmetric device compared to the symmetric device (< 170 W L^{-1}).

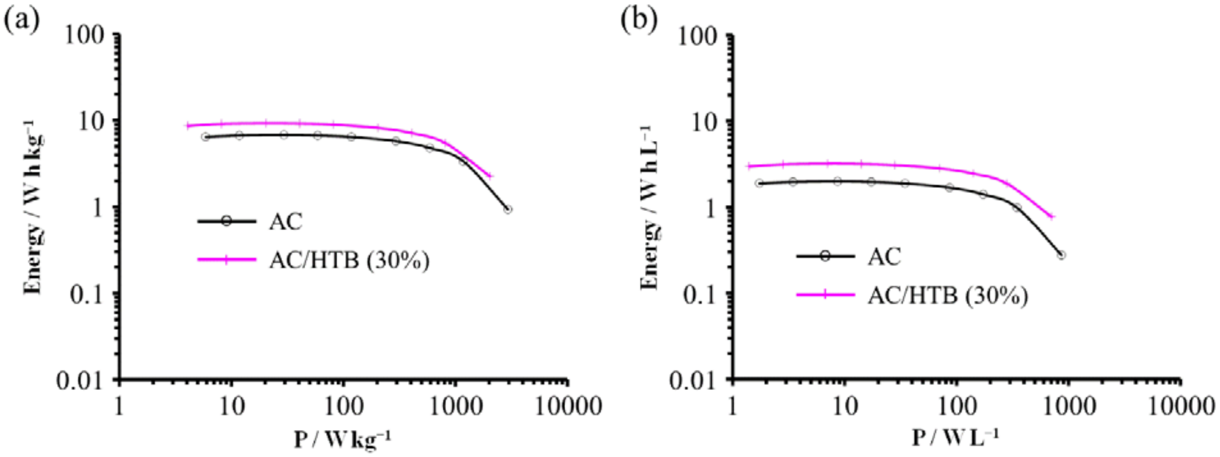

Figure S5. Ragone plot of AC/HTB (30%)/AC hybrid asymmetric device. For comparison, a symmetric device consisting of AC was plotted together. The mass of each positive and negative electrode was the same for the asymmetric and symmetric devices.

Cyclic voltammogram of negative electrode (AC) after cycle test.

After the cycle test, the negative electrode (AC) of the hybrid asymmetric device was analyzed by cyclic voltammetry in 1M H_2SO_4 at 25 °C. The measurement was conducted at a sweep rate of 1 mV s^{-1} in the potential range from −0.1 to 0.9 V (*vs*. Ag/AgCl). As clearly observed in the voltammogram, the negative electrode exhibits a distinct redox peak at the same redox potential of HTB. This result indicates that positively charged oxoammonium cations were attracted to the negatively charged electrode (AC) and then adsorbed in AC.

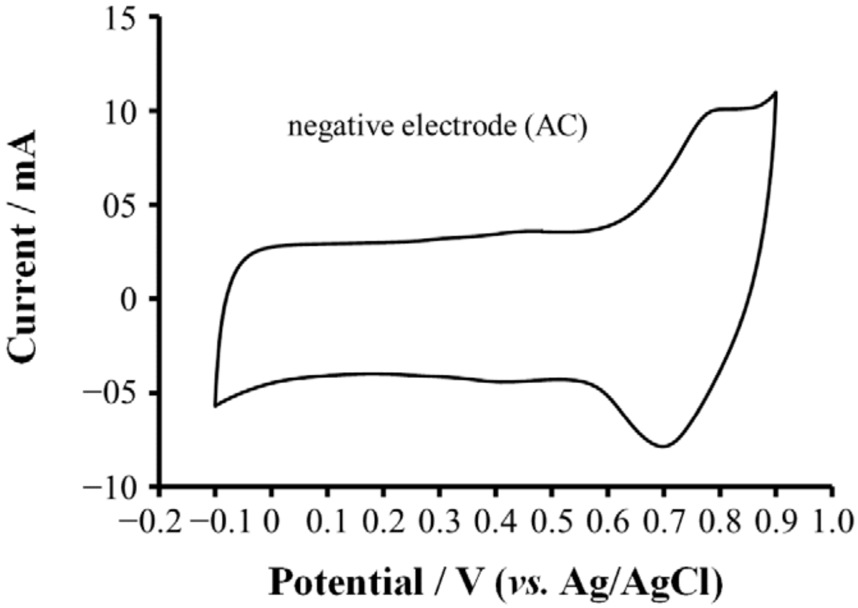

Figure S6. Cyclic voltammogram of the AC negative electrode after the cycle test by using the hybrid asymmetric device. The measurement was conducted in 1 M H_2SO_4 at a sweep rate of 1 mV s^{-1} in the potential range from −0.1 to 0.9 V.