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Energize Electrochemical Double Layer Capacitor by Introducing an Ambipolar Organic Redox Radical in Electrolyte

Lintong Hu^[a], Chao Shi^[a], Kai Guo^[a], Tianyou Zhai^[a], Huiqiao Li,^{*[a]} and Yonggang Wang^{*[b]}

Abstract: Carbon based electrochemical double layer capacitors (EDLCs) generally exhibit high power and long life, but low energy density/capacitance. Pore/morphology optimization and pseudocapacitive materials modification of carbon materials have been used to improve electrode capacitance, but leading to the consumption of tap density, conductivity and stability. Introducing soluble redox mediators into electrolyte is a promising alternative to improve the capacitance of electrode. However, it is difficult to find one redox mediator that can provide additional capacitance for both positive and negative electrodes simultaneously. Here, an ambipolar organic radical, 2, 2, 6, 6-tetramethylpiperidinyloxy (TEMPO) is first introduced to the electrolyte, which can substantially contribute additional pseudocapacitance by oxidation at the positive electrode and reduction at the negative electrode simultaneously. The EDLC with TEMPO mediator delivers an energy density as high as 51 Wh kg⁻¹, 2.4 times of the capacitor without TEMPO, and a long cycle stability over 4000 cycles. The achieved results potentially point a new way to improve the energy density of EDLCs.

EDLCs based on the symmetric carbon electrodes are attracting extensive attention, due to their outstanding power density and super-stability.^[1] However, their energy density is still much lower than rechargeable batteries.^[2] Accordingly, many efforts have been devoted to improve the energy density of EDLCs via advanced design of electrode materials.^[3-12] One typical approach is to increase the efficient surface area of carbon materials for ions adsorption,^[13-18] but which generally results in the reduction of tap density of electrodes. Another is to modify the carbon materials with pseudocapacitive materials (such as metal oxides and conductive polymers) to combine the adsorption capacitance and pseudocapacitive capacitance,^[19-22] but with the sacrifice of conductivity and stability of electrodes.

Other than the electrode materials, a new interesting approach to increase the energy density is to introduce soluble redox mediators into the traditional inert electrolyte, which can contribute additional pseudocapacitance to EDLCs through the redox reaction of the active electrolyte.^[23, 24] Recently, different redox mediators have been reported, such as halogen ions,^[25, 26] Cu/Cu²⁺,^[27] Fe²⁺/Fe³⁺,^[28, 29] and hydroquinone/quinone.^[30] However, the reported redox mediators are usually oxidized/reduced at specific potentials, which can only provide capacitance for one electrode (positive or negative) in EDLCs.

The enhanced capacitance on single electrode aggravates the asymmetric behavior of two electrodes, generally resulting in a limited voltage window and corresponding energy density.^[31-34] Hence, researchers turned to introduce dual redox mediators for two electrodes, respectively.^[35-41] However, such dual redox mediators would make the system quite complex. A highly desirable way is to find an ambipolar redox mediator that can contribute capacitance for two electrodes at one time.

Herein, we design an active-redox-electrolyte supercapacitor by introducing an ambipolar organic radical (TEMPO) to the aqueous electrolyte. As shown in Figure 1, TEMPO is a quite stable organic radical due to its resonance structure of delocalized free electrons and the steric protection of four α -methyl groups.^[42] It can lose one electron to be oxidized or obtain one electron to be reduced, which thus can energize the positive and negative carbon electrodes at the same time with largely increased capacitance. Moreover, the electron-transfer rate constant of TEMPO is approximately in the order of 10⁻² cm s⁻¹, several orders of magnitude larger than other redox mediators,^[43, 44] favoring a high potential of excellent rate performance. To assemble a full capacitor, activated carbon (AC) is used as electrode material, and aqueous electrolyte containing TEMPO is served both as posolyte and negolyte with an ion-exchange membrane in between. During charge, at the positive electrode, TEMPO is oxidized to TEMPO⁺ and the electrolyte anions are adsorbed at the interface of AC/electrolyte; simultaneously, at the negative electrode, TEMPO is reduced to TEMPO⁻ and the electrolyte cations are adsorbed at the interface of AC/electrolyte. Cations diffuse across the ion exchange membrane from positive to negative electrode while electrons transfer in the external circuit. During discharge, the opposite process occurs.

Electrolyte preparation and the synthesis of AC are given in

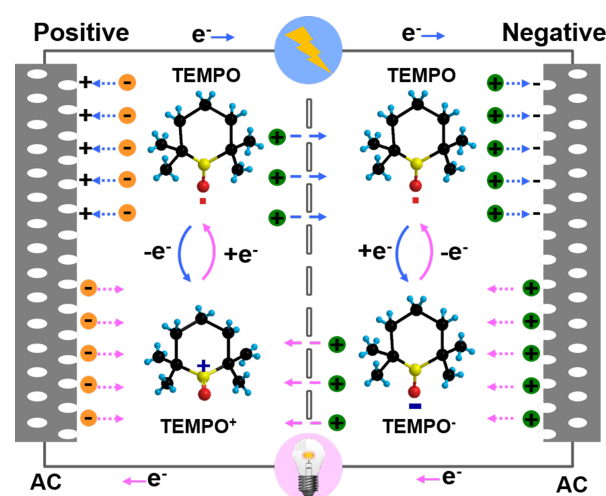


Figure 1. Schematic illustration of the operation mechanism of the supercapacitor using TEMPO as an ambipolar redox mediator.

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the experimental section. The morphology and porous structure of the prepared AC were characterized by SEM, TEM measurements and Nitrogen sorption analysis (Figures S1, S2 and S3). It is found that the AC has rich mesopores and micropores with a large surface area up to $2972 \text{ m}^2 \text{ g}^{-1}$. Raman measurement shows the partial graphitization characteristic of the AC (Figure S4). XPS analysis of the AC indicates small amount of surface oxygen functional groups (Figure S5).

Prior to assembling the full capacitor, the effect of pH value of electrolyte on the redox behavior of TEMPO was detailedly investigated by cyclic voltammetry (CV) measurements. Figure 2a shows the CV curves of AC electrode in the neutral electrolytes with and without 1 mM TEMPO. The CV curve of AC in the neutral electrolyte with 1 mM TEMPO exhibits two pairs of redox peaks located at +0.59 V/ +0.57 V and -0.2 V/ -0.38 V (vs. SCE). In contrast, the CV curve of AC without TEMPO shows a typical rectangle shape of EDLC without any humps. The two redox pairs with enough potential gap in between, indicating that TEMPO can be used as an ambipolar redox mediator. However, in the neutral electrolyte, the reversibility and response current density of the redox reaction at negative potential range are much lower than that at positive potential range. Therefore, we further investigated the two redox reactions of TEMPO separately by CV tests with different potential windows and pH values (Figure 2b). In the electrolyte with a pH value of 14 and 1 mM TEMPO, the CV curve of AC electrode shows a pair of reversible peaks at -0.47 V/ -0.44 V (vs. SCE), which is different from the rectangle-shaped CV curve presented without TEMPO (the left of Figure 2b). Furthermore, the comparison of CV curves tested in the electrolytes of pH 7 and pH 14 exhibits that the redox reaction of TEMPO in pH 14 is negatively shifted with higher response current density and reversibility, indicating the high pH value can much improve the redox performance of TEMPO in the negative potential range (-0.85 to 0 V vs. SCE, Figure S6a). Redox behavior of TEMPO at the positive potential window (0 to 0.8 V vs. SCE) was investigated in the electrolyte with a low pH value of 1.6 (the right of Figure 2b). The adopting of a low pH environment may effectively mitigate the side reaction of oxygen evolution at the positive electrode, which helps to expand the voltage window of the device. When pH decreases to 1.6, the CV curve with 1 mM TEMPO shows a pair of broadened redox peaks centered at +0.6 V (vs. SCE). The broadened reduction peaks appearing in the negative scan may rise from the reaction intermediates.^[42, 45] It is also found that the low pH environment can positively shift the oxidation of TEMPO (Figure S6b). The results shown in Figure 2a and 2b indicate that TEMPO has a typical ambipolar characteristic, which is illustrated by Figure 2c. According to above results, the ambipolar redox behavior of TEMPO much depends on the pH values of electrolytes. Therefore, the pH 14 electrolyte with TEMPO and the pH 1.6 electrolyte with TEMPO are used as the negolyte and posolyte, respectively, which can promise the better capacitance contribution to both negative and positive electrodes.

After CV measurements, the galvanostatic charge-discharge (GCD) measurements of AC electrode in the negolyte (pH 14) and posolyte (pH 1.6) were carried out to determine the capacity contribution from TEMPO. It is observed that the capacity contribution linearly increases with the concentration of TEMPO in electrolyte (Figures S7 and S8). Figure 3 presents the GCD curves of AC electrode tested in the electrolytes with and without

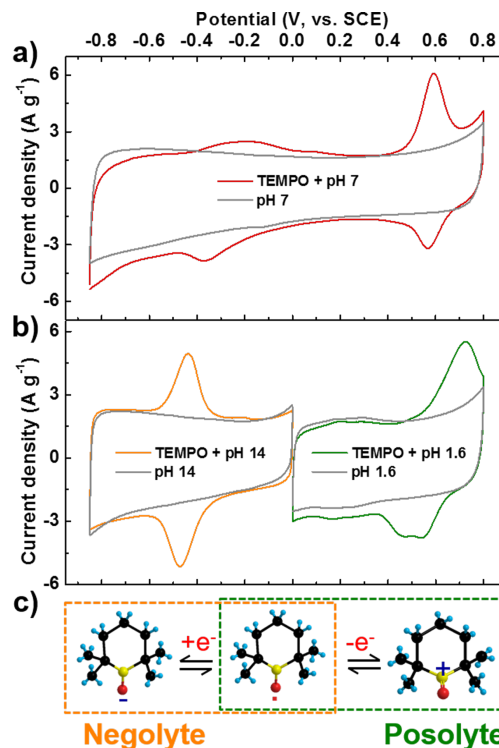


Figure 2. CV profiles of AC electrodes at 10 mV s^{-1} with and without 1 mM TEMPO in electrolyte. (a) In pH 7 electrolyte from -0.85 V to 0.8 V, (b) in pH 14 electrolyte at the negative potential window of -0.85 V to 0 V (left) and in pH 1.6 electrolyte at the positive window of 0 V to 0.8 V (right). (c) Redox reaction of TEMPO in the negative electrolyte (negolyte) and the positive electrolyte (posolyte), respectively.

10 mM TEMPO. As shown in Figure 3a and 3b, in the presence of 10 mM TEMPO, the GCD curves of AC electrode show clear potential plateaus, which is obvious different with the linear GCD curves without TEMPO. The plateaus centered at -0.45 V (Figure 3a) and +0.55 V (Figure 3b) are consistent with the redox potentials of TEMPO in the negolyte and posolyte. The rate performances of AC electrode in the negolyte and posolyte are given in Figure 3c and 3d. It can be observed the potential plateaus still exist in the GCD curves even at the high current density of 10 A g^{-1} , indicating the fast redox reaction of TEMPO. To further clarify the capacity contribution from TEMPO, the specific capacities of AC electrodes in negolyte and posolyte are also calculated based on the data shown in Figures 3c and 3d, where both the capacity (mAh g^{-1}) and current density (A g^{-1}) are normalized to the mass of AC. Rate performances of AC electrode tested in the electrolytes without TEMPO are given in the Figures S9 and S10, and the corresponding capacities are also calculated for comparison. It is demonstrated that the capacity of AC electrode in the negolyte with 10 mM TEMPO can reach up to 143 mAh g^{-1} at 1 A g^{-1} , 3.3 times of the case without TEMPO. The AC electrode in the negolyte still exhibits a capacity of 128 mAh g^{-1} at 10 A g^{-1} , 90% of the capacity at 1 A g^{-1} . In the posolyte with 10 mM TEMPO, the capacity of AC electrode at 1 A g^{-1} is 171 mAh g^{-1} , 2.7 times of the case without TEMPO. Even at a higher current density of 10 A g^{-1} , a capacity of 107 mAh g^{-1} still can be obtained in the posolyte. CV measurements at different scan rates also confirm the fast redox kinetics of TEMPO in both negolyte and posolyte (Figures S11 and S12).

To demonstrate the enhanced energy density, the negolyte

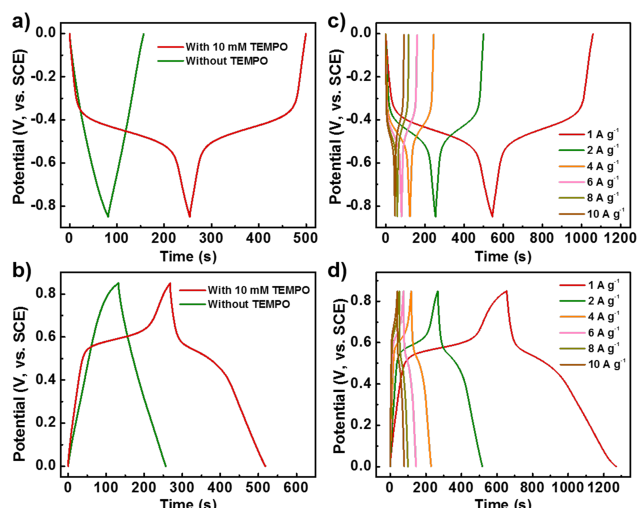


Figure 3. GCD curves of AC electrodes at 2 A g^{-1} with and without TEMPO in negolyte (a) and posolyte (b). GCD curves at different currents of AC electrodes with TEMPO in negolyte (c) and posolyte (d).

and posolyte with 10 mM TEMPO were used to build a primary capacitor with symmetric positive and negative AC electrodes, according to the illustration in Figure 1. A capacitor using the same AC electrodes with the negolyte and posolyte in the absence of TEMPO was also fabricated for comparison. Regardless of the presence and absence of TEMPO, the voltage windows of both capacitors can reach up to 1.6 V with the positive and negative potential windows of 0.8 V (Figure 4a and

4b). The GCD curve of the capacitor with TEMPO shows obvious charge/discharge plateaus, which is consistent with the potential gap between the negative and positive electrodes (Figure 4a). The GCD curve of capacitor without TEMPO shows symmetric triangle shape, indicating only the capacitive behavior (Figure 4b). GCD curves tested at different current densities of the two capacitors with and without TEMPO are gives in Figures 4c and 4d, respectively. It is clearly observed that the charge/discharge time of the capacitor with TEMPO is much longer than that without TEMPO. The capacitor with TEMPO has a maximum capacity of 60 mAh g^{-1} , nearly 2 times of that without TEMPO (Figure 4e). At 10 A g^{-1} , the capacity with TEMPO is 37.5 mAh g^{-1} , still 2 times of the case without TEMPO. The capacitor with TEMPO delivers a high energy density of 51 Wh kg^{-1} based on the total mass of AC at the positive and negative electrodes, 2.4 times of the capacitor without TEMPO (Figure 4f). The capacity of the capacitor with TEMPO can retain 96% of the initial capacity with a coulombic efficiency of $\sim 100\%$ during 4000 cycles (Figure 4g), and the GCD curves keep their shape well after cycling (Figure S13). These results indicate the introducing of TEMPO can largely increase the energy density of EDLCs. However, it is noted that the solubility of TEMPO is still low at the current stage. More efforts, for example, designing the molecule structure with $-\text{OH}$ or $-\text{NH}_2$ functional groups^[46] or choosing appropriate electrolyte such as lithium bis (trifluoromethanesulfonyl) imide,^[47] may enhance the solubility of TEMPO to further promote the energy density of EDLCs in the future.

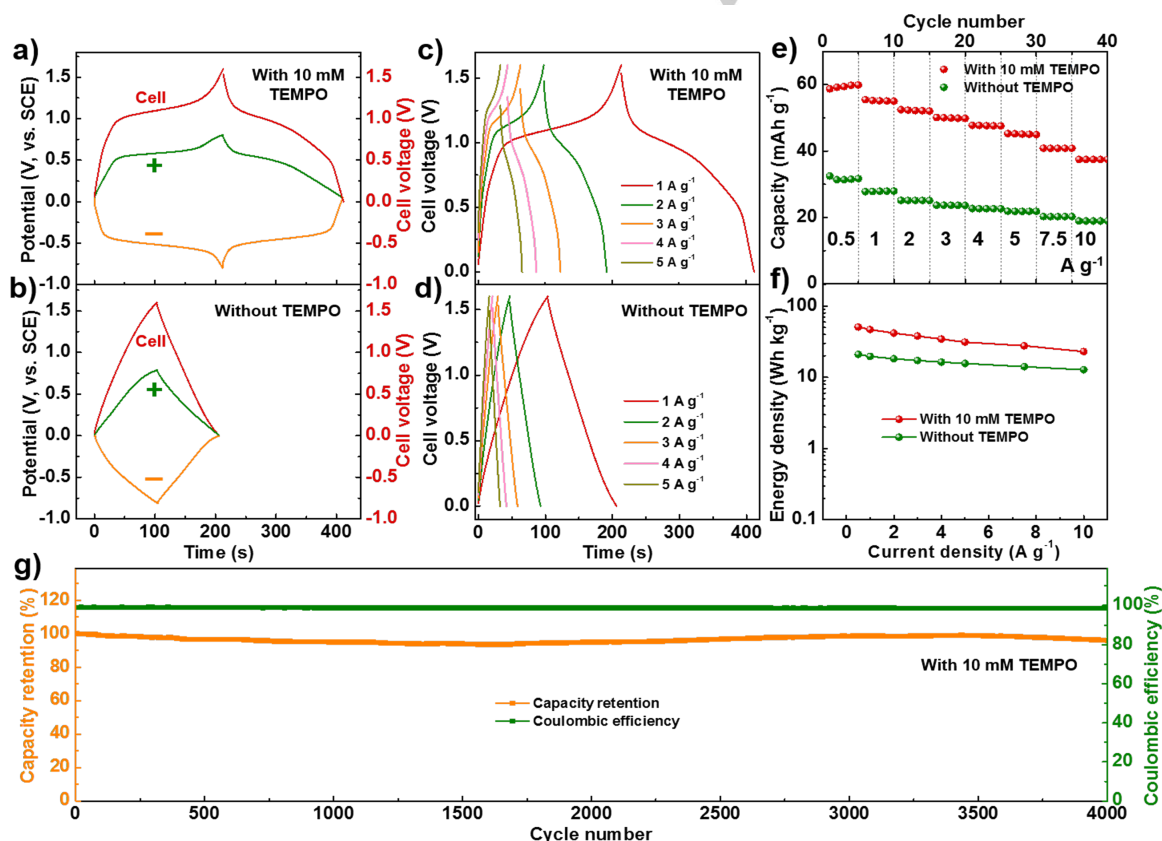


Figure 4. GCD curves at 1 A g^{-1} of the full capacitors (a) with (b) without TEMPO, along with the potentials of positive and negative electrodes. GCD curves at different current densities of the full capacitors (c) with and (d) without TEMPO. Comparison of rate performance (e) and energy density (f) of the full capacitors. (g) Cycling stability at 10 A g^{-1} of the full capacitors with TEMPO. The current density, capacity and energy density are normalized to the total mass of AC at the positive and negative electrodes.

In summary, introducing an ambipolar organic radical as the active electrolyte can effectively increase the energy density of EDLCs by its pseudocapacitance contribution to the positive and negative electrodes simultaneously. With 10 mM TEMPO in the electrolyte, the capacities of negative and positive electrodes are 143 mAh g⁻¹ and 171 mAh g⁻¹, respectively, which are about 3 times of the cases without TEMPO. The full capacitor with TEMPO delivers an energy density as high as 51 Wh kg⁻¹, 2.4 times of the cell without TEMPO, and with a long cycle stability over 4000 cycles. The achieved results potentially point a new attractive way to improve the energy density of EDLCs with little effects on electrode volume and long cycle.

Acknowledgements

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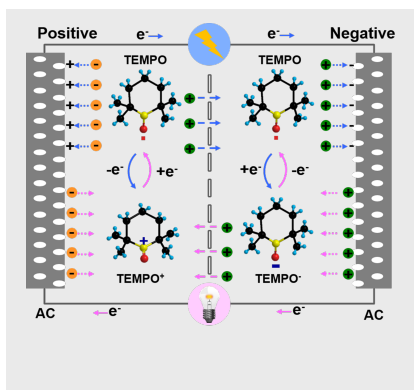
Keywords: supercapacitors • redox-active electrolyte • ambipolar redox mediator • organic radical

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Entry for the Table of Contents (Please choose one layout)

COMMUNICATION

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