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Supercapacitive properties of composite electrode consisting of activated carbon and quinone derivatives

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ABSTRACT

Physical mixing of activated carbon (AC) and a quinone derivative, 2,5-bis (pro-2-ny-1-ylamino) cyclohexa-2,5-diene-1,4-dione (coded HBU-281) was used to design a composite electrode for supercapacitors. The process proves to be simple and cost-effective; providing better performance compared to other reported methods. The electrode properties were probed in terms of composite composition, redox behavior, specific capacitance, and cycle life. The capacitance performance of the AC electrode was enhanced due to the extra redox reaction of hydroquinone/quinone couple of HBU-281. The composites recorded higher specific capacitance and excellent cycle stability than the individual electrodes (AC or HBU-281). This excellent performance can be connected to the synergistic contribution of AC facilitating the electron distribution of HBU-281, and making pronounce its redox activity. These findings led to the conclusion that physical mixing of AC and HBU-281 can be adopted to design cheap and excellent composite electrodes for supercapacitor applications.

Introduction

Supercapacitors can be classified based on their charge storage mechanism. Electrical double layer capacitor (EDLC) involves the separating of charges at the electrode/electrolyte interface while pseudocapacitors involve fast reversible faradaic reactions or combining the two mechanisms based on the nature of an electrode material. A number of literatures have discussed the development of new materials for supercapacitor electrode [1,2]. In these developments, various carbon materials have been extensively studied. These include CNTs [3], graphites [4], carbon aerogels (CAGs) [5], carbon nanofibers (CNFs) [6] and activated carbon [7]. AC due to its excellent specific surface area, high porosity, and low cost has been considered as a prospective electrode material. Nonetheless, the disadvantage associated with it is that the total surface area is not available to the ions in the electrolyte [8]. This is due to the presence of micropores (<2 nm), which decreases the formation coverage of the electric double layer. Furthermore, the capacitance of the carbon material is limited to just the double layer; thus, the capacitance of these carbon materials can be enhanced by introducing electro-active material, which is able to undergo quick and reversible redox reaction (pseudocapacitance). Metal oxides [9] have proven to be the best contributing to high capacitance, but cost and availability have been a problem for their commercial application. Conducting polymers, due to their ability to offer a continuous range of oxidation state and leading to an increase in the electrode's potential, have also gained recognition [10,11], but researches are still battling with the stability of these polymers. Quinone-based materials have attracted considerable attention as electrode materials for energy storage because they are cost effective, environmentally friendly and exhibit high reversibility [12]. They have been discovered to provide theoretical capacitance comparable to lithium ion batteries [13]. In acidic electrolyte, quinone compounds attached to carbon materials enhance capacitance through fast reversible two-electron transfer reactions [14]. Various methods have been developed on how to combine quinone species and carbon materials to enhance the charge storage performance of carbon. These include: grafting reaction via diazonium chemistry [15] and adsorption [16]. Table 1 shows a summary of some methods investigated so far; outlining the carbon support used, the working voltage and the specific capacitance. According to Ref. [20], Klemen et al. compared

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Table 1
Summary of some methods of carbon/quinone electrodes design investigated so far.

Organic compound	Carbon support	Method of preparation	Electrolyte	Voltage (V)	Scan rate/current density	Specific capacitance (F/g)	Reference	Cell type
Amino-anthraquinone	Activated carbon	Grafting reactions	0.1 M H_2SO_4	−0.5 to 0.5	10 mV s^{-1}	195	[17]	Half cell
Poly(anthraquinonyl sulfide)	Graphene	Situ polymerization	0.5 M H_2SO_4	−0.2 to 0.8	0.5 A/g	349	[18]	Half cell
Para-benzoquinone	Activated carbon	Impregnation	0.5 M H_2SO_4	−0.2 to 0.8	10 mV s^{-1}	350	[19]	Half cell
Calyx arene[4]	Carbon black or activated carbon black	Electrochemical grafting reaction	1 M $LiPF_6$ (EC/DEC)	2 to 4	–	–	[20]	Half cell
Anthraquinone	Porous carbon nanotubes	Solvothermal reaction	1 M H_2SO_4	−0.4 to 0.4	1.0 A/g	710	[21]	Half cell
Quinone	Activated charcoal	Physisorbed	1 M H_2SO_4	0 to 0.6	10 mV s^{-1}	200	[22]	Half cell
Quinone derivative	Activate carbon	Physical mixing	1 M H_2SO_4	−0.2 to 0.8	100 mV s^{-1}	205	This work	Half cell

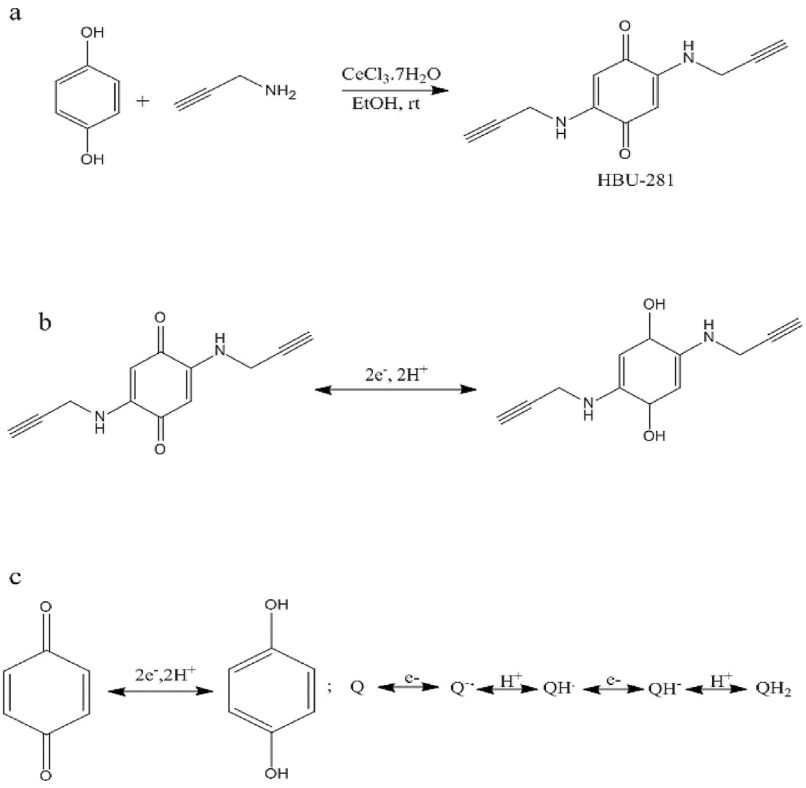
composite electrode consisting of carbon/quinone derivative via (i) physical mixing and (ii) electrochemically grafted lithium ion battery. They discovered that the cycling stability of the physically mixed composite electrode was comparable to the electrochemically grafted electrode.

In this paper, we investigate the capacitance enhancement of activated carbon by physically blending it with a quinone derivative. The properties of the electrode designed were investigated in terms of amount of quinone derivative added, redox behavior and specific capacitance against scan rate and cycle life. The potential of physically mixing quinone with carbon materials simplifies the electrode design process and makes it more cost effective and environmentally friendly.

Experimental

Synthesis of HBU-281

300 mg (2.725 mmol) of hydroquinone was dissolved in 5 ml ethanol. A solution containing 0.628 ml (11 mmol) of propargylamine and 30.7 mg (0.136 mmol) $CeCl_3{\cdot}7H_2O$ in 10 ml of ethanol was added. The reaction mixture was stirred for 12 h at room temperature. The precipitate was filtered by vacuum filtration and then washed successively with 2-propanol and hexane. The precipitate was finally dried overnight at 100 °C with a reaction yield of 95%. The resulting product was a red powder with a melting point of 300 °C. The structure of HBU-281 was investigated using ^{1}H nuclear magnetic resonance (^{1}H NMR) spectroscopy



Scheme 1. (a) Synthesis reaction of the HBU-281 from hydroquinone and propargylamine, (b) redox reaction of the HBU-281 and (c) redox reaction of quinone (Q)-hydroquinone (QH2) couple in acidic medium ([H+] > [Q]).

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