

Highly stable 2,3,5,6-tetrachloro-1,4-benzoquinone electrodes for supercapacitors



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ABSTRACT

Carbon materials have enjoyed wide applications in supercapacitors because of their high surface area which guarantees a high power output through the formation of an electric double layer (EDL). However the energy stored by this EDL mechanism is often insufficient and as such there is the need to upgrade them for higher energy applications. Quinone materials are attracting interest because of their pseudocapacitance contributions which help to boost the energy density of supercapacitors. In this study, composite supercapacitor electrodes are prepared by mechanically mixing 2,3,5,6-tetrachloro-1,4-benzoquinone (TCBQ) and activated carbon. An investigation of 5% w/w and 10% w/w of this quinolic material as a pseudocapacitance material to activated carbon in 1 M HCl aqueous electrolyte delivers a specific capacitance of 236 F g⁻¹ and 240 F g⁻¹ comparable to 190 F g⁻¹ of just activated carbon over a potential range of -0.3 V–0.9 V vs Ag⁺/Ag. Contrary to what is commonly observed, this material is highly insoluble in the electrolyte medium and remains stable with cycling, recovering 99.57% (for 10% w/w addition) and 99.13% (for 5% w/w addition) of its initial capacitance after cycling at 500 mV s⁻¹ scan rate. The findings in this report potentially provides a cheaper yet efficient route to boost the energy density of activated carbon using TCBQ for high energy supercapacitor applications.

1. Introduction

The present global energy crisis has given the search for renewable energy sources a boost in momentum. One such area that race against the demand of renewable energy source is the supercapacitor. So far various improvements are widely spread equally across all sectors from the cell system to the cell design [1–4]. As electrochemical device of great importance, the supercapacitor has been used as energy-power devices by assisting batteries at peak power and for short term power boost [5,6]. A classical electrode material for supercapacitor should possess amongst other properties a regular pore-size material with high surface area, high electrochemical and physical stability and should be very conductive [7–10]. In this regard, supercapacitors as energy-storage devices are fast charging and discharging, having high power densities, superior rate properties and incredible cycle life ability compared with batteries [11]. Carbon in its different forms is the most used commercial electrode material with specific capacitances ranging from 100 to 150 F g⁻¹ [2,12–14]. Often for carbon materials usually

used as supercapacitor electrodes, the mechanism of charge storage ideally occurs by a physical process. Due to the polarization of electrode surfaces, ions of opposite charges migrate to opposite poles which establish an electric double layer (EDLC) that stores energy [3]. Alternatively, by introducing pseudocapacitance materials like RuO₂, MnO₂, MoO₃ and electroactive conducting polymers (ECP), a resultant electron/charge transfer occurs across the electrode-electrolyte layer which increases capacitance enormously [14–16]. This process can simply be attributed to a Faradaic process occurring across the electrode-electrolyte layer of the supercapacitor. In one case when carbon was doped with nitrogen, a specific capacitance of 400 F g⁻¹ was achieved [17]. The energy stored by a supercapacitor is given by Eq. (1) as below:

$$E = \frac{1}{2}CV^2 \quad (1)$$

It therefore becomes reasonable that by increasing either the voltage range or the delivery capacitance or both of a supercapacitor component, significant additions can be made to the energy component. Quinone and its derivatives have been used by many researchers to

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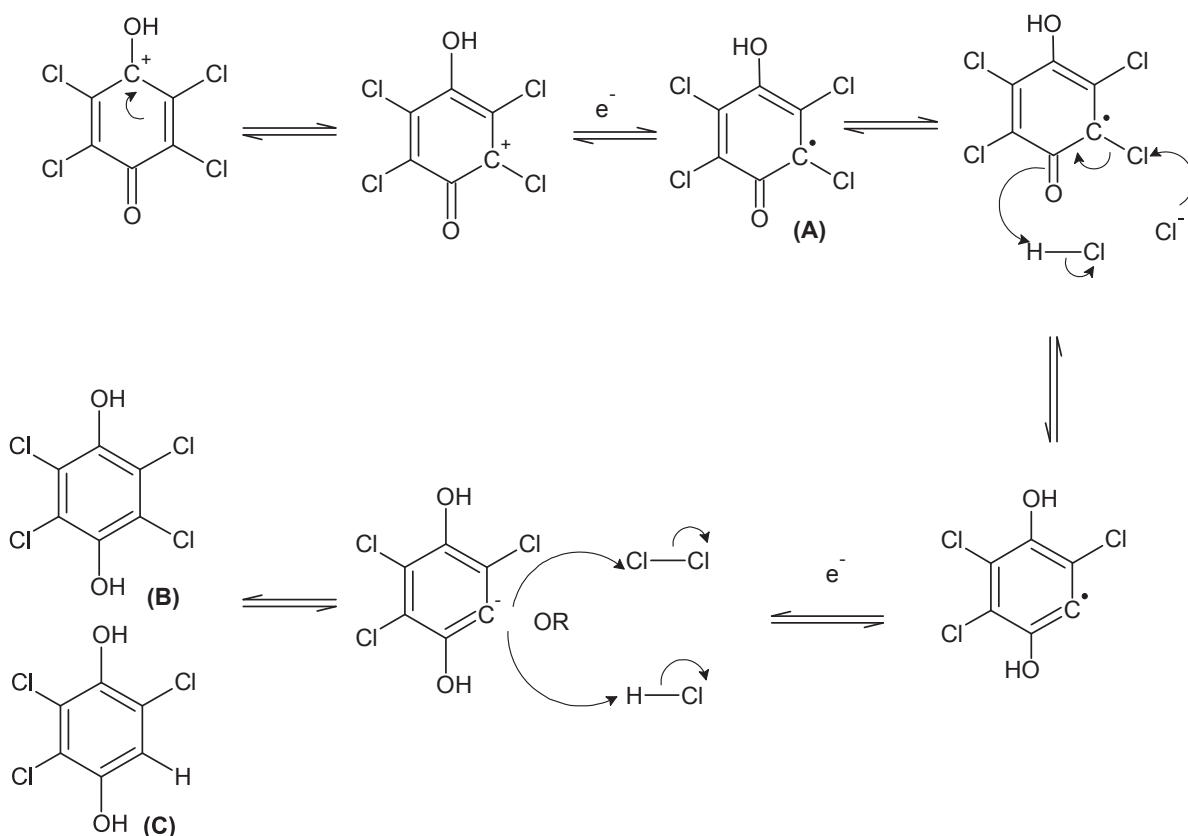


Fig. 1. Mechanism of redox reaction of 2,3,5,6-tetrachloro-1,4-benzoquinone.

boost the capacitance of EDLCs [18–21]. By a mechanism often involving several hydro/benzoquinone intermediates, these derivatives are able to transfer charges across the electrode-electrolyte interphase through a complex but yet reversible (more often than not) Faradaic reactions [18,20,21]. Generally these derivatives are either incorporated into the electrode material [18,19,21] or as redox-active electrolyte [20,22] species where they tremendously improve the capacitance by introducing a redox component. Dominica et al. by incorporating dibromodihydroxybenzene into $\text{KOH}_{(\text{aq})}$, generated a redox-active electrolyte with a capacitance value of 314 F g^{-1} using activated carbon electrodes [22]. Elsewhere, by a simple mechanism of agitation, Chandrapratap et al. physically adsorbed hydroquinone into the pores of activated charcoal which resulted in a 40% pseudocapacitance contribution to the EDLCs of activated carbon [23]. It's also been reported that by doping polypyrrole electrodes with 1,4-benzoquinone, it is able to achieve specific capacitance of 540 F g^{-1} and charge capacitance of 104 mAh g^{-1} compared to 236 F g^{-1} and 50 mAh g^{-1} of undoped polypyrrole electrodes [24].

This work discusses the pseudocapacitive contributions made by 2,3,5,6-tetrachloro-1,4-benzoquinone to activated carbon as electrode material for supercapacitors. The structure and charge storage mechanism of this material is investigated using cyclic voltammetry, galvanostatic charge/discharge test, fourier transform infrared spectroscopy and impedance measurements according to standard electrochemical techniques, over an extended voltage range. A possible reaction pathway for this material is predicted as a network of reactions taking place in the electrochemical medium which is also proposed in Fig. 1.

1.1. Materials and methodology

Activated carbon (AC) (MSC30, specific surface area of $3000 \text{ m}^2 \text{ g}^{-1}$) powder was supplied by Kansai Cokes. Poly(vinylidene

fluoride) (PVDF), 2,3,5,6-Tetrachloro-1,4-benzoquinone (TCBQ) 99%, N-methyl-2-pyrrolidone (NMP) and concentrated hydrochloric acid (HCl) were purchased from Aldrich company.

1.2. Electrode materials

In order to prepare the electrode materials, a mixture of 17:1 wt:wt and 8:1 wt:wt ratio of AC:TCBQ were first mixed in a mortar to form a uniform powder. To 9 parts of these powders, 1 part of PVDF binder in NMP solution (10 wt%) was added and mixed into a uniform slurry. The slurry was coated onto $1 \text{ cm} \times 1 \text{ cm}$ platinum electrode surface which had been previously cleaned through sonication in distilled water and acetone. The solvent was evaporated at 80°C in an oven and further dried in vacuum oven to constant weight at 100°C . Electrode coatings with an average mass of 1.4 mg cm^{-2} was recorded after drying to constant weight. The final electrodes were composed of 85:5:10 and 80:10:10 with respect to AC:TCBQ:PVDF (wt%), and thereby labeled respectively as TCBQ-5 and TCBQ-10. As a reference electrode, AC:PVDF, 90:10 was prepared through same process.

1.3. Electrochemical measurement

An Autolab PGstat 100, Eco Chemie potentiostat/galvanostat instrument was used to analyze the electrochemical properties of the electrodes. The examination was done in a 1 M HCl aq solution, by using a three electrode system with Pt-counter electrode and Ag/AgCl as the reference electrode. Potential ranges for the examination were set between -0.2 V to 0.8 V and -0.3 V to 0.9 V giving a total voltage range of 1 V and 1.2 V . Generally, the electrochemical test consisted of cyclic voltammetry (CV) test, galvanostatic charge/discharge test (C-D), electrochemical impedance and cycle test. CV test were done at scan rates of 100 mV s^{-1} , 200 mV s^{-1} , 300 mV s^{-1} , 500 mV s^{-1} and 1000 mV s^{-1} . Galvanostatic charge/discharge were conducted by

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