



A melt route for the synthesis of activated carbon derived from carton box for high performance symmetric supercapacitor applications



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HIGHLIGHTS

- Activated carbons were prepared using carton box as the precursor.
- Eutectic NaOH–KOH melt activated carbon shows better graphitization.
- Both large specific capacitance and excellent rate capability were achieved.
- The high surface area and mesoporosity is beneficial for ion adsorption and diffusion.

ARTICLE INFO

Article history:

Received 24 September 2015

Received in revised form

6 December 2015

Accepted 1 January 2016

Available online xxx

Keywords:

Activation

Micropore

Mesopore

Supercapacitor

Two-electrode system

ABSTRACT

Activated carbon materials have been playing a significant role in addressing the challenges posed with the ever-increasing demand for alternative clean and sustainable energy technologies. In the present study, a facile strategy is proposed for generating porosity in porous carbons by using carton box as the precursor and eutectic NaOH–KOH melt as the activating agent. The prepared carbon materials have a very high specific surface area up to $2731 \text{ m}^2 \text{ g}^{-1}$ with large pore volume of $1.68 \text{ cm}^3 \text{ g}^{-1}$, which is greater than that of only KOH was used at the same mass ratio. Electrochemical studies based on symmetric supercapacitor devices demonstrating that the NaOH–KOH melt activated porous carbon exhibits significantly improved rate capability in the range of $0.5\text{--}75 \text{ A g}^{-1}$ despite of the similar specific capacitance with respect to KOH activated carbon at a low current density of 0.5 A g^{-1} . The remarkable deviations in capacitive behavior at high current density for the NaOH–KOH melt activated porous carbon and KOH activated samples highlighting the specific surface area is not the only parameter that determines the capacitive performance especially at high charge–discharge rate.

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1. Introduction

Development of alternative and sustainable energy systems has gained tremendous momentum during the past decade due to the growing scarcity of fossil fuels and continually increasing greenhouse effect [1,2]. Supercapacitors, also known as electrochemical capacitors, have emerged as promising candidates for energy conversion and storage due to their merits of high power capability, outstanding cyclic stability, simple principle, and speedy dynamics of charge propagation over other storage devices [3–5]. Currently, carbonaceous materials become the most popular materials for supercapacitors owing to their high surface area and structural diversity. To date, a lot of efforts have been devoted to the synthesis

of novel carbon materials with diverse structures including nanotubes [6], onions [7], aerogels [8], templated carbons [9], carbide-derived carbons [10], graphene [11], and hierarchical porous carbons [12], for supercapacitor applications. Although these carbon-based materials can afford better capacitive performance than that of commercial activated carbon, their relatively high costs and somewhat tedious synthesis processes is a serious issue for their widespread application. Up to now, only activated carbon (AC) has been commercially used as supercapacitor electrode material because these materials not only offer a very large surface area for double layer formation, but their synthesis is also relatively facile and cost-effectiveness [13]. The main challenges facing activated carbon-based supercapacitors are low energy density and the decreasing availability of fossil-based carbon source. Considering the demand for sustainable eco-friendly resources and process, it is essential to be able to produce AC with renewable sources or

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recycled carbon-containing wastes. For example, activated carbon obtained from various renewable biomass precursors such as coffee beans [14], tree leaves [15], sucrose [16], corn grains [17], bamboo [18], and others have been explored to generate activated carbon on a laboratory scale for using in supercapacitor electrodes. The random tortuous and bottle-neck pores in activated carbons, however, dramatically slow down the ion transfer from the electrolytic solution to the inner surface especially at high current loads, which severely limit their practical applications as high-performance energy storage devices [19,20]. Strategies for improving the performance of supercapacitors including increasing the conductivity, constructing a hierarchical structure, introducing mesoporosity, and reducing the dimensions of carbon have been developed to improve their power capability without deteriorating their high energy density and cycle life [21]. Unfortunately, the capacitance and energy density are often unsatisfactory at high charge–discharge rate that still cannot meet the higher requirements of electric systems, ranging from portable electronics to hybrid electric vehicles. Thereupon, it is urgent and significant to search novel, environmentally-friendly, low-cost, and high-performance carbon-based supercapacitors with excellent capacitive performance.

At present, physical (e.g., air, CO₂ and steam) and chemical (e.g., H₃PO₄, ZnCl₂ and KOH) methods of activation processes are well-known strategies to produce highly porous carbon materials with a large surface area from coal derived precursors or organic compounds, which have been widely adopted in the recent years [22,23]. Compared to physical activation, chemical activation has superior advantages such as lower activation temperatures and higher surface area although it has disadvantages including the corrosiveness of the chemical agents and the necessary washing process [23]. Among the various chemical reagents, KOH is widely used since it can result in AC with defined micropore size distribution, and a very high specific surface area of up to 3000 m² g⁻¹ [23,24]. Their poor electrical conductivity and the predominant tortuous pores, though results in the transfer limitation of electrolyte ions during rapid charge–discharge processes, deteriorating its rate capability [25–28]. Exploiting of NaOH as an activating agent is less referred in the literature, although it has some advantages over KOH such as low price, simple handling procedure, and less corrosive behavior [29,30]. Recently, by means of NaOH as the activating agent, porous carbon materials with higher mesopore volume and better conductivity than those prepared with KOH were produced [29,31]. The larger mesopore volume and conductivity made the NaOH activated carbon as promising electrode materials for high performance supercapacitors especially at high charge–discharge rate. Nevertheless, NaOH activated porous carbons usually have a low surface area due to the difference reaction mechanisms as compared with KOH, which will give a low capacitance [32]. Inspired by the above points, it is reasonable to applying NaOH and KOH composites as a new activating agent for generating more mesopore volume in carbons while maintaining the good conductivity. Furthermore, the melting points of pure NaOH and KOH are both above 300 °C (323 °C for NaOH and 404 °C for KOH), the eutectic points for particular mixtures of these elements, NaOH/KOH = 51.5:48.5 is only about 165 °C [33]. The very low eutectic points made the NaOH/KOH molten salt is one of the promising approaches for obtaining various materials at lower temperatures as compared with conventional high temperature solid-state reactions [33]. Recently studies suggested that the activation mechanism of carbon and KOH starts with solid–solid reactions and then proceeds via solid–liquid reactions including the reduction of potassium (K) compound to form metallic K, the oxidation of carbon to carbon oxide and carbonate, and other reactions among various active intermediates [22,23,27]. Therefore,

the low eutectic points of NaOH–KOH melt can lead to generate liquid phase at relatively low temperature, and the subsequently diffusion of the liquid will increase the interfacial area for carbon and activation agent that can improve the utilization of activation agent as identified by the characteristic of the solid–liquid reactions [34–36]. Therefore, NaOH–KOH melt would be an effective activating agent to generate the pore network in carbons, although the use of molten-salt as an alternative to obtain porous carbon materials has not received much attention yet.

As the prosperous development of express delivery industry, carton box have become an essential part of our modern lifestyle because of its low cost and abundant production. Nevertheless, in most cases, carton box was discarded or open burning without effective utilizations, recycling or reuse is the best current solution for reducing the disposal problem. Lee and coworkers reported the activated carbon with surface area of 416 m² g⁻¹ and the maximum specific capacitance of 160 F g⁻¹ from waste newspapers by pyrolysis and KOH treatment technique [37]. Very recently, high surface area porous carbon was synthesized from waste office paper by a two-step process which includes hydrothermal treatment followed by high temperature KOH activation [38]. In the present work, we demonstrate the porous carbon synthesized from the waste carton box as the electrode material for supercapacitor applications with low amount NaOH–KOH melt as the activating agent. The as-prepared porous carbon materials have a very high specific surface area up to 2731 m² g⁻¹ with large pore volume of 1.68 cm³ g⁻¹, which is greater than that of only KOH was used at the same mass ratio. Electrochemical studies based on symmetric supercapacitor devices demonstrating that the NaOH–KOH melt activated porous carbon exhibits significantly improved rate capability with respect to KOH activated carbon over a large range of current densities.

2. Experimental section

2.1. Synthesis of porous carbon

The waste carton box was used as the precursor for the synthesis of the porous carbon material. The material was synthesized by a two-step process involving pre-carbonization followed by chemical activation. The carton box was used without any pre-processing. Briefly, the carton box was cut into small pieces and carbonized at 600 °C in a tubular furnace for 2 h, and the as-prepared carbonized sample named pretreated carton box (PCB). Then, the as-prepared PCB was divided into three groups (I, II and III). The chemical active processes were performed under high purity nitrogen atmosphere with a fixed heating rate of 5 °C min⁻¹. The furnace was allowed to cool to ambient temperature naturally. The product obtained after activation was washed with 1 M HCl and plenty of water to remove the inorganic impurities then drying under vacuum at 70 °C for 24 h. Group I was mixed with NaOH–KOH (NaOH/KOH = 51.5:48.5, mass fraction) with mass ratios of NaOH–KOH/PCB at 1/1, 2/1 and 3/1, followed by activation at 800 °C for 1 h under continuous N₂ flow. The obtained samples were named NKAC-1, NKAC-2 and NKAC-3. Group II was mixed with KOH with mass ratios of KOH/PCB at 1/1, 2/1 and 3/1, then activated at 800 °C for 1 h. The resulting products were denoted as KAC-1, KAC-2 and KAC-3, respectively. Group III was mixed with NaOH with mass ratios of 3/1, and activated at 800 °C for 1 h, and the final product was named as NAC.

2.2. Sample characterization

The powder X-ray diffraction (PXRD) measurements were performed by using Shimadzu X-ray 6000 diffractometer with Cu-Ka1

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