



High-energy green supercapacitor driven by ionic liquid electrolytes as an ultra-high stable next-generation energy storage device

Ranjith Thangavel^a, Aravindaraj G. Kannan^b, Rubha Ponraj^b, Vigneysh Thangavel^c, Dong-Won Kim^b, Yun-Sung Lee^{a,*}

^a Faculty of Applied Chemical Engineering, Chonnam National University, Gwang-ju 500-757, Republic of Korea

^b Department of Chemical Engineering, Hanyang University, Seoul 04763, Republic of Korea

^c Department of Electrical and Electronics Engineering, Institute of Aeronautical Engineering, Hyderabad 500 043, Telangana, India

HIGHLIGHTS

- Hierarchical porous carbon is obtained from waste watermelon rind.
- The biomass carbon exhibits high surface area along with micro and mesopores.
- A high voltage supercapacitor is constructed with ionic liquid electrolyte.
- The supercapacitor delivers high energy (174 Wh kg^{-1}) and high power (20 kW kg^{-1}).
- The supercapacitor exhibits remarkable stability (150,000 cycles) at 60°C .

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ABSTRACT

Development of supercapacitors with high energy density and long cycle life using sustainable materials for next-generation applications is of paramount importance. The ongoing challenge is to elevate the energy density of supercapacitors on par with batteries, while upholding the power and cyclability. In addition, attaining such superior performance with green and sustainable bio-mass derived compounds is very crucial to address the rising environmental concerns. Herein, we demonstrate the use of watermelon rind, a bio-waste from watermelons, towards high energy, and ultra-stable high temperature green supercapacitors with a high-voltage ionic liquid electrolyte. Supercapacitors assembled with ultra-high surface area, hierarchically porous carbon exhibits a remarkable performance both at room temperature and at high temperature (60°C) with maximum energy densities of $\sim 174 \text{ Wh kg}^{-1}$ (25°C), and 177 Wh kg^{-1} (60°C) – based on active mass of both electrodes. Furthermore, an ultra-high specific power of $\sim 20 \text{ kW kg}^{-1}$ along with an ultra-stable cycling performance with 90% retention over 150,000 cycles has been achieved even at 60°C , outperforming supercapacitors assembled with other carbon based materials. These results demonstrate the potential to develop high-performing, green energy storage devices using eco-friendly materials for next generation electric vehicles and other advanced energy storage systems.

1. Introduction

The ever increasing global energy demand, the reduced availability of fossil fuels and rising environmental concerns have shifted the focus of energy research toward the development of sustainable and renewable energy sources such as solar and wind energy [1,2]. However, these energy sources are highly intermittent in nature and require grid-scale energy storage devices for back-up and load-leveling during peak production and consumption periods, respectively. Moreover, the energy-intensive automotive sector requires electric vehicles with low

CO_2 emissions. To meet the demands of these next-generation applications, a significant improvement in the energy density and cycle life of current energy storage devices is crucial [3]. However, the current state-of-the-art lithium ion batteries have almost reached their practical limit [4,5]. Although sodium ion batteries are being developed due to their low cost, their energy density is lower than that achievable in lithium ion batteries. Specifically, problems related to low specific power and poor cycle life limit the practical use of these battery systems [6,7]. On the other hand, supercapacitors are highly promising candidates for next-generation applications due to their high power densities

* Corresponding author.

E-mail address: leey@chonnam.ac.kr (Y.-S. Lee).

and long cycle life [8,9].

Supercapacitors have been receiving increased interest due to their high power density and long cycle life. They can work as an independent energy source or complement different energy sources such as batteries, fuel cells, solar cells and wind power, where quick bursts of power are required [3,10]. Supercapacitors are generally classified as non-faradic electrical double layer type charge accumulation at the porous carbon electrode-electrolyte interface, and pseudocapacitors based on reversible faradaic redox surface charge storage in metal oxides/sulfides [9,11–14].

Supercapacitors based on double layer formation are always an eminent choice for commercial devices due to their simple construction and long cycle life than pseudocapacitors. Porous carbon-based materials have been widely used in double layer type supercapacitors due to their high surface area, which provides large number of favorable sites for double layer formation [15,16]. Recently, supercapacitors exhibiting high energy densities closer to those of rechargeable batteries and power densities much higher than those of batteries have been reported [17,18]. Moreover, the energy density of supercapacitors can be further increased by extending the operating potential using ionic liquid electrolytes and high surface area carbon architecture [19–21]. However, supercapacitors employing ionic liquid electrolytes suffer from relatively poor cycling performance and failure at high power due to the highly viscous nature of the ionic liquid. This can be overcome by employing a high surface area carbon with mesoporous hierarchical architecture and favorable functional groups that could aid penetration of ionic liquids into deeper pores and get stored for a longer time [20,22].

Activated carbons, the widely used candidate of choice for double layer type supercapacitors deliver unsatisfactory performance with highly viscous and low diffusivity ionic liquids [23,24]. The highly microporous conventional activated carbon cannot provide space for large sized ions in ionic liquids, curtailing the performance of supercapacitor device [25–27]. Various forms of carbon with wider pores have been extensively explored including graphene, carbon nanofibers, carbon nanotubes, template-derived carbon and porous carbon [28–32]. Although graphene and template-derived carbons are considered to be more promising, there are major challenges that impede the practical application of these materials including graphene restacking during processing, poor control over pore distribution and the tedious process required for template preparation [33,34].

Bio-mass, which is widely an agricultural waste has recently emerged as an alternate source for porous carbon and has been adapted in energy storage devices [35,36]. Bio-mass derived porous carbon can achieve ultra-high surface area than graphene, along with tailored architecture, large pore volume, mesoporous morphology, tuned surface chemistry and wide commercial viability [37,38]. In addition, porous carbon produced from bio-wastes can provide a sustainable solution due to its renewable nature, low cost and wide availability [32,39]. The major research involves investigation of bio-mass derived porous carbon towards low energy aqueous supercapacitors, lacking exploration in high performing - high energy supercapacitors.

Herein, we report the application of highly mesoporous carbon derived from waste watermelon rind (hereafter referred as CWM) with a hierarchical pore structure and controlled pore morphology towards high energy supercapacitors driven by high voltage ionic liquid electrolyte. Watermelon rind is a bio-waste that constitutes about 33% of watermelon material (*Citrullus lanatus*). The worldwide production of watermelons is more than 100 million tons per year and waste watermelon rind are an inexpensive, and abundantly available carbon precursor that could enable the possibility of sustainable and green high energy – high power next generation devices. Although, porous carbon from watermelon peels have been investigated for supercapacitor applications, the most of the research focuses on capacitor working with aqueous electrolytes with a limited working potential [29,36–38]. Additionally, the true and practical merits with symmetrical two electrode

testing has not been widely investigated. In this research, we evaluate the performance of such symmetrical supercapacitor working in an ionic liquid electrolyte under a wide working potential (4V) and at elevated temperature. The current research establishes the possibilities of utilizing sustainable, high energy storage systems for advanced applications.

2. Experimental

2.1. Synthesis of CWMs

Watermelons used in this work were purchased from local markets in Korea, and the rind was collected after removing the edible portions. The rind was rinsed with DI water and dried in an oven at 120 °C, and it was then powdered prior to the carbonization step. Carbonization was carried out at 600 °C in an Ar atmosphere. Chemical activation with KOH was used to generate pores, and the ratio of carbon to KOH was controlled to be 1:5. After activation in a tube furnace at 800 °C for 2 h in an Ar atmosphere, the product was washed with a 0.1 M HCl solution, followed by rinsing with water and ethanol several times. The material was then dried and used for further characterization and supercapacitor performance evaluation.

2.2. Material characterization

The morphologies of the CWM were characterized using scanning electron microscopy (SEM, JEOL JSM 6701F) equipped with energy dispersive spectroscopy (EDS) and high-resolution transmission electron microscopy (HRTEM, JEOL, JEM 2100F). X-ray diffraction (XRD, Rigaku D/MAX 2500 diffractometer) was used to characterize the crystalline structure of CWM. X-ray photoelectron spectroscopy (XPS, VG Multilab ESCA System, 220i) was used to confirm carbonization and to characterize the surface functional groups. Nitrogen adsorption/desorption isotherms were recorded using an ASAP-2020, Micromeritics, USA apparatus at 77 K. The powder samples were degassed at 300 °C for 2 h under vacuum prior to the measurements. The micropore size distribution were determined based on non-local density functional theory (NLDFT) assuming a slit pore model. The mesopore distribution was determined based on Barrett-Joyner-Halenda (BJH) model. Raman spectra were obtained using a Dongwoo Optron, MonoRa 780i spectrometer. The electrical conductivity of the carbon powders were determined in four pin probe apparatus (Loresta-GP MCP-T610, Mitsubishi chemical analytech) at 25 °C using a carbon pellet (1 cm diameter) pressed at 10 MPa. The contact angle measurement of the electrodes with ionic liquid electrolyte was measured on Kruss DSA100 Goniometer.

2.3. Electrochemical characterization

The electrodes for supercapacitor were formulated with accurately weighed amounts of active material (80 wt%), Ketjen black (10 wt%) as conductive carbon, and Teflonized acetylene black binder (10 wt%). The electrode was pressed over a stainless-steel mesh with a mass loading of 2.5–3.5 mg cm⁻² and dried at 160 °C for 4 h in a vacuum oven. The thickness of the working electrode is calculated to be ~70–75 μm. Supercapacitors were assembled in a standard CR2032 coin cell with two symmetrical electrodes, which were separated by a porous polypropylene separator (Celgard 3401) and filled with 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide (EMImTFSI). EMImTFSI was purchased from Chem Tech Research Incorporation and was used after drying under vacuum at 100 °C for 24 h. The water content in EMImTFSI after drying was determined to be less than 20 ppm by Karl Fisher titration using a Mettler-Toledo Coulometer. All the cells were assembled in an argon filled glove box. The specific capacitance of each electrode is calculated based on the formulae C_s ($F g^{-1}$) = $2(I \times t)/(V \times m)$, where I is the applied current (A), t is the

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