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Single Step, Direct Pyrolysis Assisted Synthesis of Nitrogen-Doped Porous Carbon Nanosheets Derived from Bamboo wood for High Energy Density Asymmetric Supercapacitor

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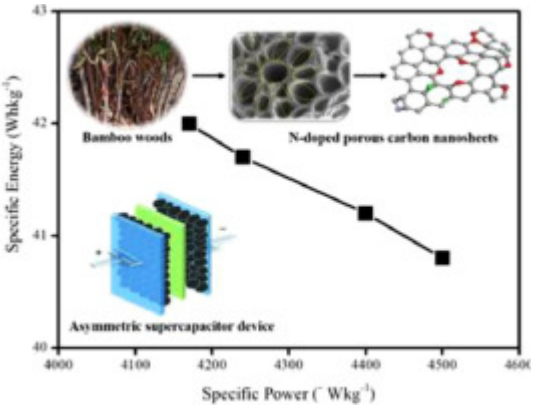
Highlights

- The porous nitrogen-doped carbon nanosheets was prepared from bamboo biomass in single-step direct pyrolysis technique.
- The nitrogen-doped carbon nanosheets exhibits a better half-cell specific capacitance than undoped carbon char.
- The highest half-cell specific capacitance of as-prepared nitrogen-doped carbon nanosheets reached 475 Fg^{-1} at 1Ag^{-1}.
- An asymmetric supercapacitor based on this nitrogen-doped carbon showed a high energy density (42 $Whkg^{-1}$ at 4.5 $kWkg^{-1}$).

Abstract

Herein, we report a scalable, large-area synthesis of nitrogen-doped carbon nanosheets (N-AC) from bamboo biomass using KOH and urea as the activating and doping agent, respectively, for high performance asymmetric supercapacitor applications. The carbonization of the biomass bamboo at 300 °C for 2 h yields N-AC_0 (char) while activation at 900°C for 2 h under inert atmosphere yields N-AC. The detailed characterization of N-AC revealed the significance of heteroatom doping for the supercapacitor application. The N-AC electrode delivered the half-cell specific capacitance of 475 Fg^{-1} at 1 Ag^{-1}. The assembled asymmetric N-AC||N-AC_0 supercapacitor device delivered the highest specific capacitance of 296 Fg^{-1} at 1 Ag^{-1} with specific energy of 42 $Whkg^{-1}$ at specific power of 4500 Wkg^{-1}, which are very high compared to other reported nitrogen-doped carbon materials. This enhanced performance can be attributed to the synergistic effect of larger specific surface area (of 769.714 m^2g^{-1}) and hetero-atom (N) doping, which helped in rapid charge-transfer, improved electrical conductivity and efficient electrode-electrolyte interactions. Furthermore, the N-AC asymmetric supercapacitor device works until 1.2 V with 6 M KOH as with 150% capacitance retention over 10,000 charge-discharge cycles. Thus, the strategy presented here provides new directions to synthesize low cost, green, sustainable electrodes for high energy supercapacitor applications.

Graphical Abstract



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Introduction

In recent decades, there has been an increase in demand for energy storage and

distribution to fulfil the expanding global energy requirements. Supercapacitors (SCs) are one of the most popular energy storage devices because they are clean energy storage devices with high specific power, quick charge-discharge rates, long cycle life, and little maintenance. Further, they overcome the defects between the traditional capacitors and batteries [1,2]. The active electrode materials are chosen based on their charge-discharge mechanisms. There are two types of charge-discharge processes: the electrostatic electrical double-layer capacitive process and the electrochemical pseudo-capacitive process. Electrical-double layer behaviour appears in carbon-based materials, whereas pseudocapacitive behaviour appears in transition-metal oxides and conducting polymers. Carbon-based electrode materials include benefits such as natural abundance, user-friendliness, greater specific surface area, electrical conductivity and thermal stability. However, they have low intrinsic capacitance with low energy density, restricting their use in practicability [3,4]. Hence, modifying the structure of the carbonaceous materials can overcome the above disadvantages [5].

Biomass-derived carbon materials such as carbon flakes [6], mesoporous carbon [7], fibers [8], carbon nanosheets [9], sponge aerogels [10], and activated carbon [11] have gained attention as the electrode materials for the SC applications [12]. Compared with the traditional sources of carbon, biomass-based precursors are the pursuit of low-cost and environmentally friendliness. The most commonly used method for improving the electrochemical performance of carbon material is the doping technique with the heteroatoms like boron, nitrogen, phosphorous, and sulfur [13]. Doping of these heteroatoms into carbon skeleton contributes to the pseudocapacitive properties in addition to the EDLC behaviour. The presence of N-active sites transfers more electrons, thereby significantly improving the electrical property of the carbon matrix. Nitrogen-rich precursors can be utilised to make nitrogen-doped carbon.

Synthesis of nitrogen-doped carbons via high-pressure hydrothermal techniques [14], higher temperature annealing [15], and other chemical methods [16, 17] have been reported in the literature. However, low-cost, scalable techniques for delivering excellent capacitive properties are yet to be explored. When nitrogen-doped carbon is employed as electrode material in the asymmetric supercapacitor, the specific energy and power can be significantly enhanced.

In this work, asymmetric supercapacitor device is fabricated by employing the prepared nitrogen-doped carbon from biomass bamboo (N-AC) as the negative electrode and the carbonized char (N-AC_0) as the positive electrode. . Bamboo is one of the fastest-growing wood, evergreen, and perennial plant distributed from plains to hills. BB contains various compounds such as cellulose, hemi-cellulose, lignin, proteins, amino acids, fats, carbohydrates that are rich in nitrogen is used for the

preparation of N-AC [18]. During pyrolysis, these compounds induce a porosity that enhances the surface area, which contributes to the improved charge storage property of the material. Thus, we demonstrated a simplified ASC device employing the prepared nitrogen-doped (N-AC) carbon as the negative electrode. To the best of our knowledge, this is the first-time fabrication of the ASC device with N-AC as a negative electrode which is prepared via direct pyrolysis of bamboo biomass.

Section snippets

Materials and Reagents

Bamboo woods biomass was procured from local supermarket. Potassium hydroxide (KOH)-99.9% pure, dilute hydrochloric acid, conducting carbon (ethylene black)-99.9% pure, ethanol, polyvinylidene fluoride (PVDF), N-methyl-pyrrolidone (NMP) were procured from Sigma Aldrich and are employed for the electrode synthesis, fabrication and electrochemical measurements. De-ionised (DI) water was collected from the Millipore system (resistivity:18 Ω) and s used for the experiments All the reagents were of…

Physiochemical Analysis

The surface morphology of the N-AC was investigated using FESEM and HRTEM. With the inclusion of urea and KOH during the activation process, noticeable surface morphological changes were observed. At the initial stage, the urea is converted to the graphitic carbon nitride (g-C_3N_4), which is then decomposed as a nitrogen-rich compound at 900^o C and finally gets doped into carbon framework [23]. **Fig.2(a, b)** depicts the FESEM images of N-AC, which has wrinkled nanosheets with porous morphology.…

Conclusion

To summarize, the N-AC porous nanosheets have been synthesized from bamboo-biomass by one-step carbonization and activation technique, towards supercapacitor application. The N-AC electrode material delivered an excellent half-cell specific capacitance of 475 Fg^{-1} at 1 Ag^{-1} with high potential window up to -1.2 V. The produced N-AC electrode material's enhanced electrochemical performance is owing to the synergistic impact of nitrogen doping and KOH activation. The highest specific capacitance…

CRediT authorship contribution statement

Sivagaami Sundari Gunasekaran: Conceptualization, Data curation, Methodology, Writing – original draft. **Arthi Gopalakrishnan:** Conceptualization, Data curation, Methodology, Writing – original draft. **Raghu Subashchandrabose:** Conceptualization, Data curation, Methodology, Writing – original draft. **Sushmee Badhulika:** Conceptualization, Funding acquisition, Investigation, Project administration, Resources, Supervision, Writing – review & editing.…

Declaration of Competing Interest

There are no conflicts to declare.…

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