



Highly compressible three-dimensional graphene hydrogel for foldable all-solid-state supercapacitor

Xianbin Liu^{a,*}, Shuai Zou^a, Kaixi Liu^a, Chao Lv^a, Ziping Wu^a, Yanhong Yin^a, Tongxiang Liang^a, Zailai Xie^b

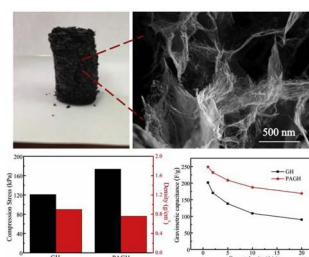
^a School of Materials Science and Engineering, Jiangxi University of Science and Technology, 86 Hongqi Road, Ganzhou 34100, China

^b College of Chemistry, Fuzhou University, Fuzhou, Fujian 350018, China

HIGHLIGHTS

- A novel three-dimensional graphene hydrogel mediated by phytic acid is prepared.
- The PAGH possess 3D network structure with low density and enhanced compressive strength.
- The PAGH electrode exhibits excellent electrochemical performance.
- All-solid-state supercapacitor delivers outstanding flexibility.

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:

High compressive strength
Graphene hydrogel
High energy density
All-solid-state supercapacitor

ABSTRACT

The fabrication of three-dimensional (3D) graphene-based macroscopic materials with superior mechanical and electrical properties for flexible energy storage devices is still extremely challenging. Here, we report a novel 3D graphene hydrogel decorated by the biomass phytic acid (PAGH) with developed porosity and strengthen mechanical property via hydrothermal and freeze-drying methods. The phytic acid molecules are intercalated into the graphene sheets, enabling robust network structure. This induces the formation of materials with larger specific surface area, lower density and enhanced compressive strength compared with pure GH. When directly employed as an electrode, the PAGH exhibits a high specific capacitance of 248.8 F g^{-1} at 1 A g^{-1} and excellent rate performance of 67.9% as current density increasing to 20 A g^{-1} . Furthermore, the all-solid-state supercapacitor based PAGH can deliver outstanding cycle life (86.2% after cycling 10,000 times), glorious energy density (26.5 Wh kg^{-1}) and power density (5135.1 W kg^{-1}). The prepared device shows stable electrochemical behaviors at random bending angles. Therefore, the present work will open a new avenue to design and fabricate new flexible and portable graphene-based electrodes for future applications in energy storage devices.

1. Introduction

Searching of safe, flexible and highly efficient energy storage devices is crucial important to cope with the rapid development of portable electronic equipments pervasive in modern society [1–6]. Supercapacitors are expected to be one of the most prominent devices owing to their high power density and long cycling life. Supercapacitors can

be classified into two types according to their mechanism of energy storage: electrochemical double-layer capacitors (EDLCs) and pseudocapacitors [7–10]. In EDLCs, the energy is physically stored by the accumulation of charged species on the surface of electrodes. The properties of this kind of supercapacitors are primarily contingent on the features of the electrode materials. The study of EDLCs electrode materials has mostly focused on carbon materials, such as fullerene

* Corresponding author.

E-mail addresses: xianbin_liu@jxust.edu.cn, liuxianbin1987@126.com (X. Liu).

[11–13], carbon nanotubes [14–16] and active carbon [17–19]. However, the present carbon-based electrodes only show mediocre energy density and power density, limiting their extensive application. Therefore, the development of new carbon materials with high energy density and power density is of great significance and remains challenge.

Graphene, a single layer of carbon atoms patterned in a hexagonal lattice, has recently demonstrated great potential as a new EDLCs electrode material because of its high intrinsic electrical conductivity, excellent mechanical flexibility, exceptionally large theoretical surface area (up to $2630 \text{ m}^2 \text{ g}^{-1}$), and theoretical gravimetric capacitance of approximately 550 F g^{-1} [20–22]. However, experimental graphene only exerts an inferior and unsatisfactory gravimetric capacitance ($< 200 \text{ F g}^{-1}$) because of the strong π - π interaction between graphene sheets, which leads to a decrease in the accessible surface area. To overcome these issues, many efforts have been centered on the development of graphene-based macrostructures with a three-dimensional (3D) porous network [23,24]. Typical example includes graphene hydrogel (GH) consisting of interconnected 3D porous network which is an ideal electrode material for EDLCs applications [25,26]. However, the neat GH has nonnegligible drawbacks of structural collapse and destroys of intrinsic texture easily under compression because of their brittle skeletons and strong inter-sheet attractive forces [27–29]. Currently, surface functionalization of graphene by organic molecules has been verified effective to regulate the mechanical properties and 3D porosity [30]. For example, Jin-Yong Hong et al. reported that the embedding of polyvinyl alcohol molecule could improve the mechanical durability and increase the gravimetric and volumetric capacitances [28]. Yang Zhao et al. reported that the form of graphene could be mediated by polypyrrole leading to high highly compression-tolerant properties [31]. However, the aforementioned method requires pre-synthesis of polymer, which is either quite complex or enormous time/energy consuming. Thus, a sustainable and green method for developing functional graphene with 3D porosity and enhanced mechanical property is urgently needed.

Phytic acid (PA), an abundant natural product found in plants, has six phosphate groups [32–34]. These groups provide a great deal of viable cross-linking sites that may “stitch” graphene sheets to form a 3D macrostructure. Herein, we innovatively use PA as reducing and skeleton molecules to prepare a PA-mediated graphene hydrogel (PAGH) for supercapacitor electrode *via* hydrothermal method accompanied by freeze-drying treatment. The fabrication process is facile and straightforward as depicted in Scheme 1. The PAGH presents a hierarchical network structure with high specific surface area up to $637 \text{ m}^2/\text{g}$ and robust mechanical properties with compression strength of 171 kPa. Attributed from these functional groups, the PAGH could be directly used as a high-performance electrode in supercapacitor. The current electrochemical test indicates that the specific capacitance reaches up to 248.8 F g^{-1} at a current density of 1 A g^{-1} . With the current density

increasing to 20 A g^{-1} , the specific capacitance still remains at 67.9% of the initial value. To further explore the application of PAGH, we also constructed a PAGH based all-solid-state supercapacitor. The obtained supercapacitor delivers impressive cycle life, energy density, and power density. Meanwhile the favorable flexibility was demonstrated by investigating the electrochemical properties at various bending angles.

2. Experimental

2.1. Materials

Flake graphite powder was supplied by JinRiLai Graphite co., LTD (Qingdao, China). Phytic acid was purchased from Aladdin Industrial Corporation (Shanghai, China). Other reagents were purchased from Sinopharm Chemical Reagent Company (Shanghai, China).

2.2. Preparation of PAGH

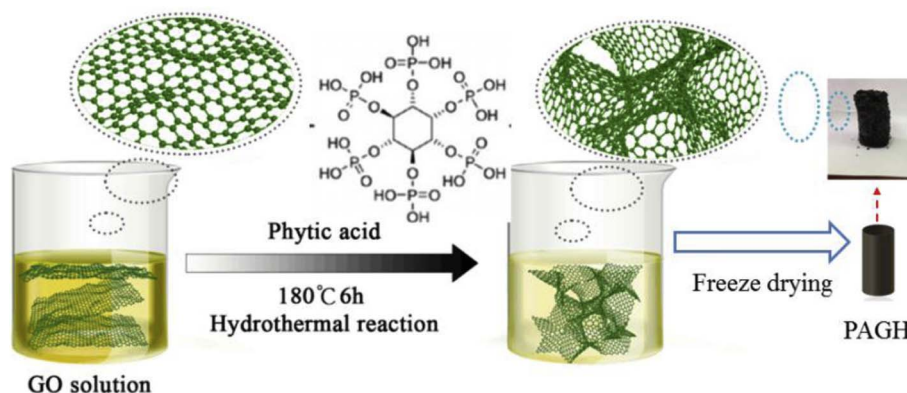
First, graphene oxide (GO) was prepared from the flake graphite using an improved Hummers method [35]. Then, an aqueous GO solution (2.0 mg mL^{-1}) and PA (5 mL) were mixed in a beaker. The mixed solution was transferred into a Teflon[®] reactor, and reacted at 180°C for 6 h. The obtained wet sample was soaked and washed with plenty of deionized water. Finally, the sample was freeze-dried to obtain the PAGH. The GH was prepared by the same procedure without PA.

2.3. Characterization

Transmission electron microscopy (TEM) was performed on a Tecnai G2F20 (FEI, USA). Scanning electron microscopy (SEM) analysis was accomplished using a SUPRA 55 (ZEISS, Germany) with secondary electron at 10 kV. The electrical conductivity was measured by a four-probe method using compressed pellets with a SDY-4 four-point probe meter (China). The BET surface area was measured using the nitrogen adsorption-desorption isotherm on a NOVA 2000e surface area and pore size analyzer (USA). The crystalline structure was characterized using an Ultima III X-ray diffractometer (XRD, Rigaku, Japan). Fourier transform infrared spectroscopy (FTIR) and Raman spectroscopy analysis were recorded with a Nicolet 5700 (Thermo, USA) and Invia (Renishaw, UK), respectively. The elemental composition was analyzed on an ESCALAB 250 X-ray photoelectron spectroscope (XPS, Thermo Scientific, USA). The mechanical properties were characterized by a CMT-5505 electron universal testing machine (SANS, China).

2.4. Electrochemical measurement

The electrochemical analyses of obtained samples were carried out in a three-electrode system equipped with an Ag/AgCl reference electrode and a platinum counter electrode in a $1 \text{ M H}_2\text{SO}_4$ aqueous



Scheme 1. The fabrication process of the 3D macroscopic PAGH.

Download English Version:

<https://daneshyari.com/en/article/7725543>

Download Persian Version:

<https://daneshyari.com/article/7725543>

[Daneshyari.com](https://daneshyari.com)