



# Excellent electrochemical performances of cabbage-like polyaniline fabricated by template synthesis



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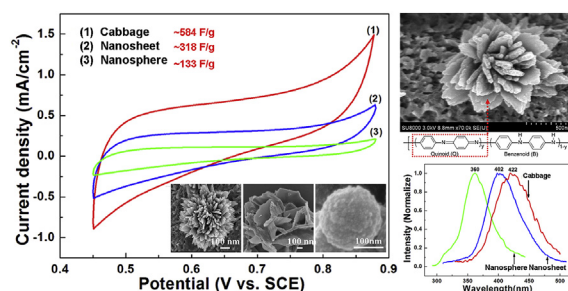
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## HIGHLIGHTS

- A novel route to fabricate cabbage-like polyaniline (PANI) by polymeric template.
- The specific capacitance of the PANI is 584 F/g and it also shows high stability.
- The good performance of the PANI is attributing to larger-scale  $\pi$ - $\pi$  conjugated system.

## GRAPHICAL ABSTRACT



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## ABSTRACT

In this article, we explore a novel route to fabricate cabbage-like polyaniline (PANI) by in situ polymerization of aniline using the hydroxylated poly (methyl methacrylate) nanospheres (i.e. PMMA-O-H-NS) as a template. A maximum specific capacitance of 584 F/g (the current density is 0.1 A/g) is achieved at 10 mV s<sup>-1</sup> as well as a high stability of over 3000 cycles (the decrease in the SC is ~9.1%), which suggests the potential application of the cabbage-like polyaniline in supercapacitors. The predominant electrochemical performances of the cabbage-like polyaniline can be attributed to their large surface area and larger-scale  $\pi$ - $\pi$  conjugated system present in the quinoid structure of the PANI molecular chain, which can drastically facilitate electron diffusion and improve the utilization of the electroactive PANI during the charge/discharge processes. Accordingly, the facility of charge transfer can decrease resistance along with the PANI molecular chain to improve the electrochemical stability and achieve high-capacitance response characteristics. The present study introduces a new synthesis method for the development of various morphology of other conducting polymer, which may find potential applications in a variety of high-performance electrochemical devices.

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## 1. Introduction

Presently, energy storage remains one of the great challenges due to the depletion of fossil fuel resources. Among the optional major contenders for power source, supercapacitors, fuel cells, and

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lithium ion batteries are being strongly considered in recent years [1–3]. Supercapacitors, also called electrochemical capacitors (ECs), have attracted considerable interest in energy storage due to its possession of higher energy density than dielectric capacitors and higher power density than batteries [4]. Supercapacitors have been extensively used for power buffer applications in cameras and mobile phones, electric vehicles for transportation and digital communication, etc [5]. It is well-known that transition-metal oxides, carbon materials and conjugated conducting polymers can be investigated for their use in supercapacitors. However, the toxicity of transition metal and high cost of carbon materials (such as carbon nanotube, fullerene and graphene) limit their application in supercapacitors. Meanwhile, conjugated conducting polymers have been regarded as promising pseudocapacitive materials because it is easy to form large-area devices and their energy gaps and ionization potentials can be readily tuned by chemical modification of the polymer chain [6]. In addition, they can be generally produced at a lower cost than noble metal oxides and their performance is comparable to that of carbon fiber materials. Polyaniline (PANI) is one of the most studied conducting polymers [7]. It has been extensively studied as supercapacitor electrode materials because of its ease of synthesis, good environmental stability and high conductivity [8–13].

Currently, the PANI used in supercapacitors usually has good electrochemical properties with a specific capacitance of 233–1220 F/g [14]. This is because the accessible surface area of the as-prepared PANI plays an important role in influencing the participation of the conducting polymer in the charge storage process and their specific capacitance. Commonly, in order to enhance the performance of the electrode, reduce the diffusion length and facilitate the ionic motion, the porous or hollow-structured materials are synthesized so as to enhance surface-to-volume ratio and reduce the distance for mass and charge transport [15–17]. Therefore, the advent of methods for designing smaller scale and hierarchically-structured PANI nanomaterials with high specific surface area, high electrical conductivity, and a fast ion diffusion process are of great importance to further increase the capacitive performance of the as-prepared products. Accordingly, materials such as PANI nanofibers [18], nanorods [19], nanowires [20], nanotubes [21], hollow sphere [22], nanobelts [23], octahedral morphology [24], etc. have been widely studied with the expectation that such materials will have the advantages of low dimensional systems and organic conductors. In this article, we report a simple approach to prepare well-controlled cabbage-like polyaniline through a template chemical method by controlling the appropriate condition. The research reported here focuses on developing an uncomplicated chemical route for the preparation of cabbage-like polyaniline and examining the electrochemical properties of cabbage-like polyaniline, in order to provide an understanding of the new morphology of PANI, which scientists could use to find its potential applications in supercapacitors. The prepared cabbage-like polyaniline are characterized by scanning electron microscope (SEM), Raman spectroscopy, Ultraviolet–visible (UV–vis) spectroscopy and fluorescence spectroscopy. These cabbage-like polyaniline are expected to make a significant contribution to the advancement of chemical pseudocapacitor technology due to their high pseudocapacitive behavior and electrochemical stability.

## 2. Experimental

### 2.1. Materials

Methylmethacrylate (MMA), potassium persulfate ( $K_2S_2O_8$ , 99.5%), ammonium persulfate ( $(NH_4)_2S_2O_8$ , APS), aniline,

tetrabutylammonium perchlorate (TBAP) and 3-(Trimethoxysilyl)-propyl methacrylate (MATS) were purchased from Aladdin Chemistry Co., Ltd (Shanghai, China). These chemicals and other reactants were analytical grade and used without further purification except aniline, which was distilled twice under reduced pressure.

### 2.2. Preparation of hydroxylated poly (methyl methacrylate) nanospheres (i.e. PMMA–OH–NS)

The poly (methyl methacrylate) nanospheres (i.e. PMMA-NS) were prepared as follows: 80 ml deionized water and 3.7 ml MMA monomer were added into the reactor which was adequately stirred with a magnetic stirrer at 80 °C under a nitrogen atmosphere for 20 min, and then 20 ml, 4.9 mM  $K_2S_2O_8$  aqueous solution was added to the reactor and allowed to react for 4 h at 70 °C [25]. The PMMA–OH–NS were prepared as follows: The 50 ml PMMA-NS dispersed solution was diluted to 100 ml by adding deionized water into the reactor at room temperature, then 0.36 ml MATS and 0.02 g  $K_2S_2O_8$  was dropped into the reactor at 80 °C and allowed to react for 2 h with stirring under a nitrogen atmosphere. The final products were dried at room temperature for one week, then washed with alcohol three times, and subsequently dried in a vacuum oven. The SEM images and Raman spectra of as-prepared samples are shown in Figs. S1 and S2 of the Supporting Information.

### 2.3. Template synthesis of cabbage-like polyaniline

In a typical experiment, 0.1 g PMMA–OH–NS powder was added to a two necked round bottomed flask containing 25 ml water and dissolved with the aid of ultrasonication for 30 min at room temperature. Double distilled aniline (1.0 ml) and 1.0 mol/L  $(NH_4)_2S_2O_8$  solution (5 ml) were then added and mixed with stirring. The reaction was allowed to proceed at 25 °C for 36 h with continuous stirring. The dark green precipitate was then isolated by filtration, washed many times with methanol and dried under reduced pressure at room temperature. To remove the PMMA–OH–NS template, the as-prepared PANI was dispersed in appropriate volume of tetrahydrofuran for 12 h and then the final product was isolated by filtration, washed many times with a methanol/water solution and dried in air at room temperature.

### 2.4. Electrochemical performance measurement

The electrochemical performance was measured by using a conventional three-electrode configuration using a saturated calomel electrode (SCE) as the reference electrode, a platinum sheet as the counter electrode, and the Fluorine-doped Tin Oxide (FTO)-coated target mixture as working electrodes. The target mixture containing 85 wt% cabbage-like polyaniline, 10 wt% conducting carbon black, and 5 wt% polytetrafluoroethylene (PTFE 60% dissolved in  $H_2O$ , and used as a binder) was well mixed and then pressed onto the FTO (the cabbage-like polyaniline loading is ~0.12 mg). The used electrolyte was 1 M aqueous  $H_2SO_4$  solution. The experiments were performed on a computer-controlled CHI 660C electrochemical workstation (CH Instruments, Shanghai Chenhua Inc., and China).

### 2.5. Characterization

The SEM images of the cabbage-like polyaniline were obtained with a Hitachi SU8000 Field-emission Microscope (Hitachi LTD, Tokyo, Japan). A Renishaw inVia Laser Micro-Raman Spectrometer (New Mills, UK) was used to record the Raman spectra. The spectra were measured at 532 nm excitation wavelength with a resolution

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