Flexible planar asymmetric supercapacitor using synthesized few-layer graphene and activated carbon from biomass for wearable energy storage**

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We have fabricated a flexible planar asymmetric supercapacitor demonstrating high energy storage capability that can be utilized to power various flexible and wearable electronic devices. A locally available cheap source of biomass—banana peel—was used for synthesising carbonaceous materials—few-layer graphene and activated carbon—for the device. Few-layer graphene was synthesized by heating banana peel at high temperature under an inert atmosphere followed by crushing with mortar and pestle. Activated carbon was synthesized by heating banana peel impregnated with KOH at high temperature. The device was fabricated using a low-cost screen printing technique to create the current collector followed by deposition of active electrode materials and sandwiching filter paper soaked in gel electrolyte in between the electrodes. Devices showed high areal capacitance of 88 mF/cm^2 at 10 mV/s scan rate; and were satisfactory under multiple electronic cycling (100 cycles) and bending conditions. The devices can be economically fabricated on a large scale and used for developing emerging flexible electronics.

Keywords: high areal capacitance, screen printing

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1. Introduction

Recently the demand for wearable electronics has considerably increased and is expected to grow further in the future. However, practical applications are limited due to the lack of wearable energy sources. Lithium-based batteries are predominantly used in modern-day electronics but they have limitations, such as risk of explosion or electrolyte leakage, which makes them unfit for wearable applications. Hence the need has arisen for safe, environmentally friendly, low cost, thin and mechanically flexible energy storage devices, which would store energy and supply it to wearable devices. Such storage devices represent a significant paradigm shift in consumer electronics since they eliminate the necessity for carrying bulky energy sources. Hybrid supercapacitors are a promising alternative to hitherto developed energy sources for powering wearable electronics, especially sensors. The hybrid supercapacitor combines the advantages of an electric double layer capacitor (EDLC) and psuedocapacitance [1,2]. We here describe the fabrication of carbon-based, thin, mechanically flexible planar asymmetric supercapacitors using a screen printing technique for creating the current collector, incorporating synthesized few-layer graphene and activated carbon from banana peel, and transition metal oxides as the electrode material. This flexible supercapacitor can be incorporated into wearable electronics in the biomedical, smart textile, computing and processing domains. Such devices can also be utilized for environmental parameter monitoring applications [3].

2. Materials and device fabrication

2.1 Synthesis of activated carbon

Locally available cheap biomass such as banana peel (from *Musa acuminata*) was used as a precursor. The peel was washed several times with distilled water and left overnight in a vacuum oven at 80 °C to remove excess water. A small quantity (3 g) of KOH flakes was mixed with deionized (DI) water (50 mL) to prepare an aqueous solution. A small quantity (3 g) of the dried peel was added to this KOH solution and constantly stirred at 200 rpm with a magnetic bar for 2 h. The peel was then taken out from the solution and again left overnight in the vacuum oven at 80 °C to remove excess water. The KOH- impregnated peel was then heated at 600 °C for 1 h in a muffle furnace. The resulting carbonaceous material was washed multiple times with dilute HCl and distilled water until the pH reached neutrality (pH 7) and finally heated in the vacuum oven at 80 °C overnight to yield the activated carbon. It was characterized using a Brunauer–Emmett–Teller (BET) surface area analyser (Quantachrome model: Autosorb-IQ MP) and a field emission scanning electron microscope (FESEM) (Zeiss model: Sigma). The FESEM image (Fig. 1) reveals a well-developed porous structure, with a typical pore size of few hundred nm. The BET surface area was 62.03 m^2/g [4].

2.2 Synthesis of few-layer graphene

Banana peel was also used for synthesizing few-layer graphene. Thoroughly washed and dried peel was further washed with ethanol and acetone and dried at room temperature. The peel was then heated in a tubular furnace at 950 °C for 2 h under argon. The carbon residue was crushed in a mortar and pestle, resulting in few-layer graphene fragments (Fig. 2a). Raman spectroscopy (Fig. 2b) verified the presence of few-layer graphene [5].

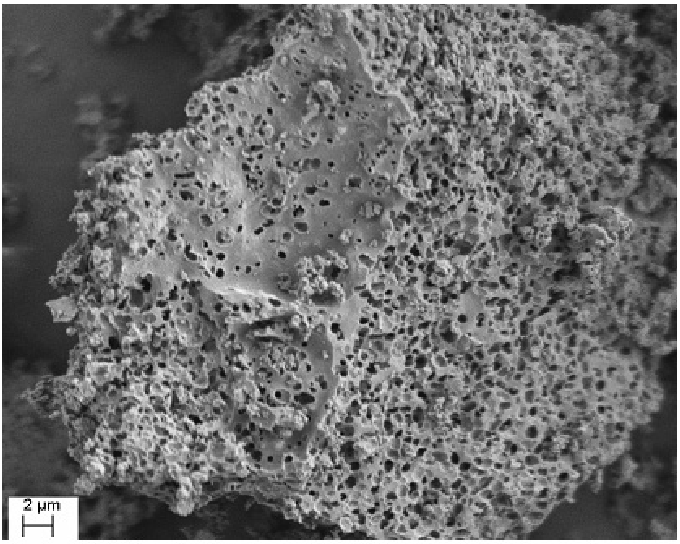

Figure 1: Field emission scanning electron microscopy (FESEM) image of activated carbon at 5000 times magnification.

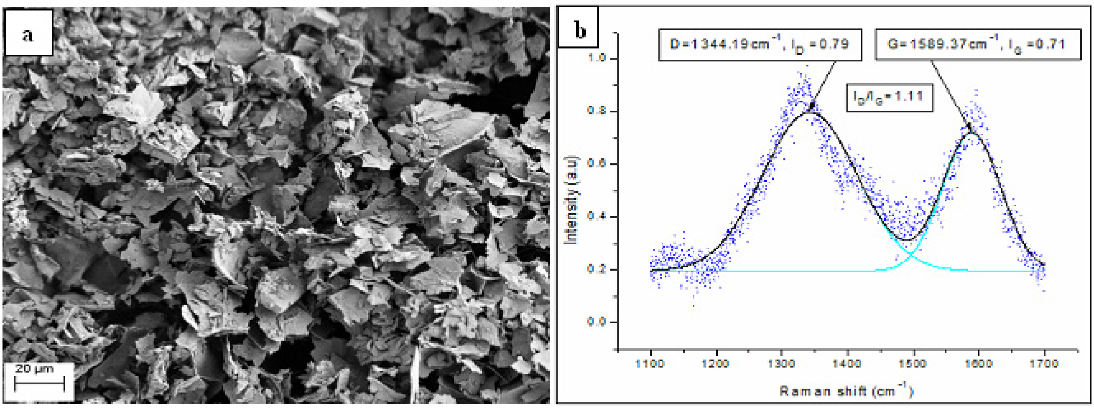

Figure 2: (a) FESEM image of few-layer graphene at 1000 times magnification; (b) Raman spectrum of few-layer graphene at 633 nm.

2.3 Fabrication of planar asymmetric supercapacitor

Silver ink current collectors were screen-printed on flexible polyethylene terephthalate (PET) substrate. Equal amounts (1 mg) of few-layer graphene (FLG) and activated carbon (AC) were sprinkled on one of the printed current collectors forming the anode. Equal amounts (0.5 mg) of NiO and Co_3O_4 along with 1 mg of few-layer graphene were sprinkled on the other printed current collector forming the cathode [6]. PVA/H_3PO_4 gel electrolyte was prepared by mixing polyvinyl alcohol (PVA) in DI water (1:10 w/w) under constant stirring at 90 °C followed by addition of 1 mL of phosphoric acid [7]. Filter paper soaked in gel electrolyte was sandwiched between the two asymmetric electrodes as shown in Fig. 3(a), with front and side views of the fabricated supercapacitor shown in Fig. 3(b,c). The anode electrode material (FLG/AC) was examined by FESEM, showing the pore sizes of the activated carbon to be in a range of 100–200 nm, and the presence of few-layer graphene flakes.

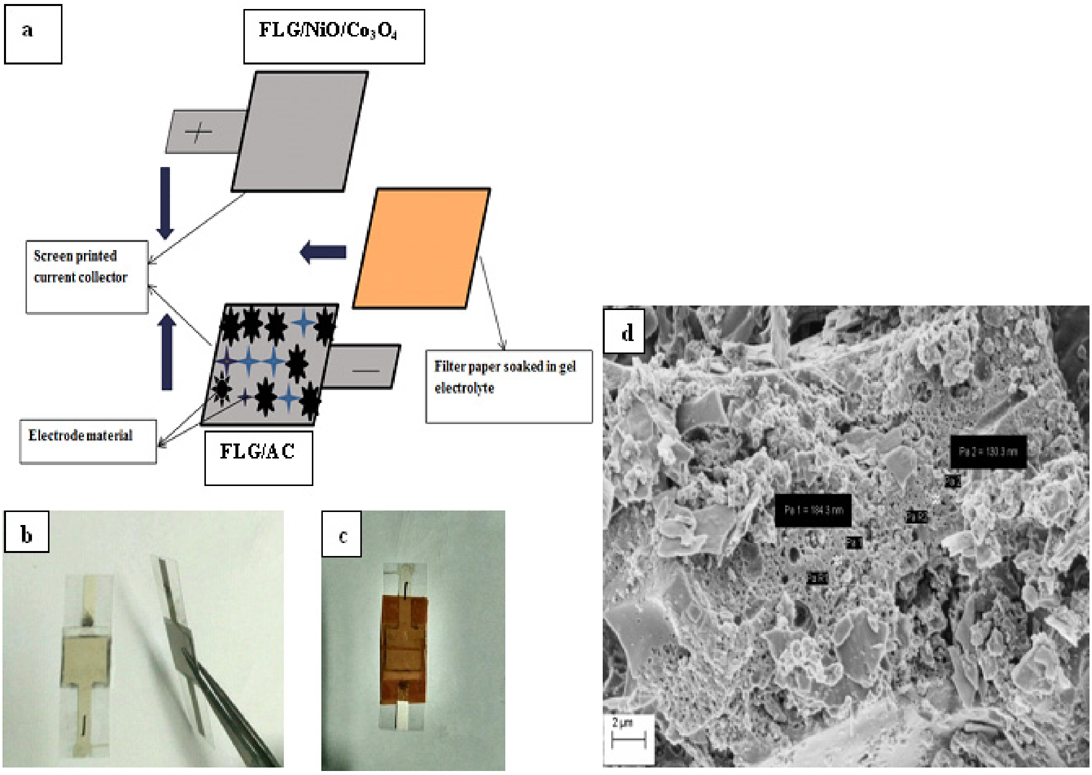

Figure 3: (a) Schematic description of fabrication of planar asymmetric supercapacitor; (b) Optical image of fabricated supercapacitor; (c) planar supercapacitor covered with sticky tape; (d) FESEM of FLG/AC at 10,000 times magnification.

3. Device characterization

The fabricated supercapacitor was tested by cyclic voltammetry (CV) at scan rates ranging from 10 mV/s to 200 mV/s (Fig. 4a). The areal capacitance C was calculated by:

$$C = A / (a\, v\, V) \tag{1}$$

where A is the area of the CV curve, a is the area of the active part of the device (3.61 cm^2), v is the scan rate in mV/s and V is the potential window (1 V) (Fig. 4b) [8]. A lifecycle test was done by carrying out 100 CV cycles at a scan rate of 100 mV/s; the supercapacitor retained 36% of its initial capacitance (Fig. 4c). A series-connected planar supercapacitor were tested under unbent and bent conditions and a 133% increase in capacitance was observed in the bent condition, which may be due to better interaction between electrode and electrolyte (Fig. 4d,e,f).

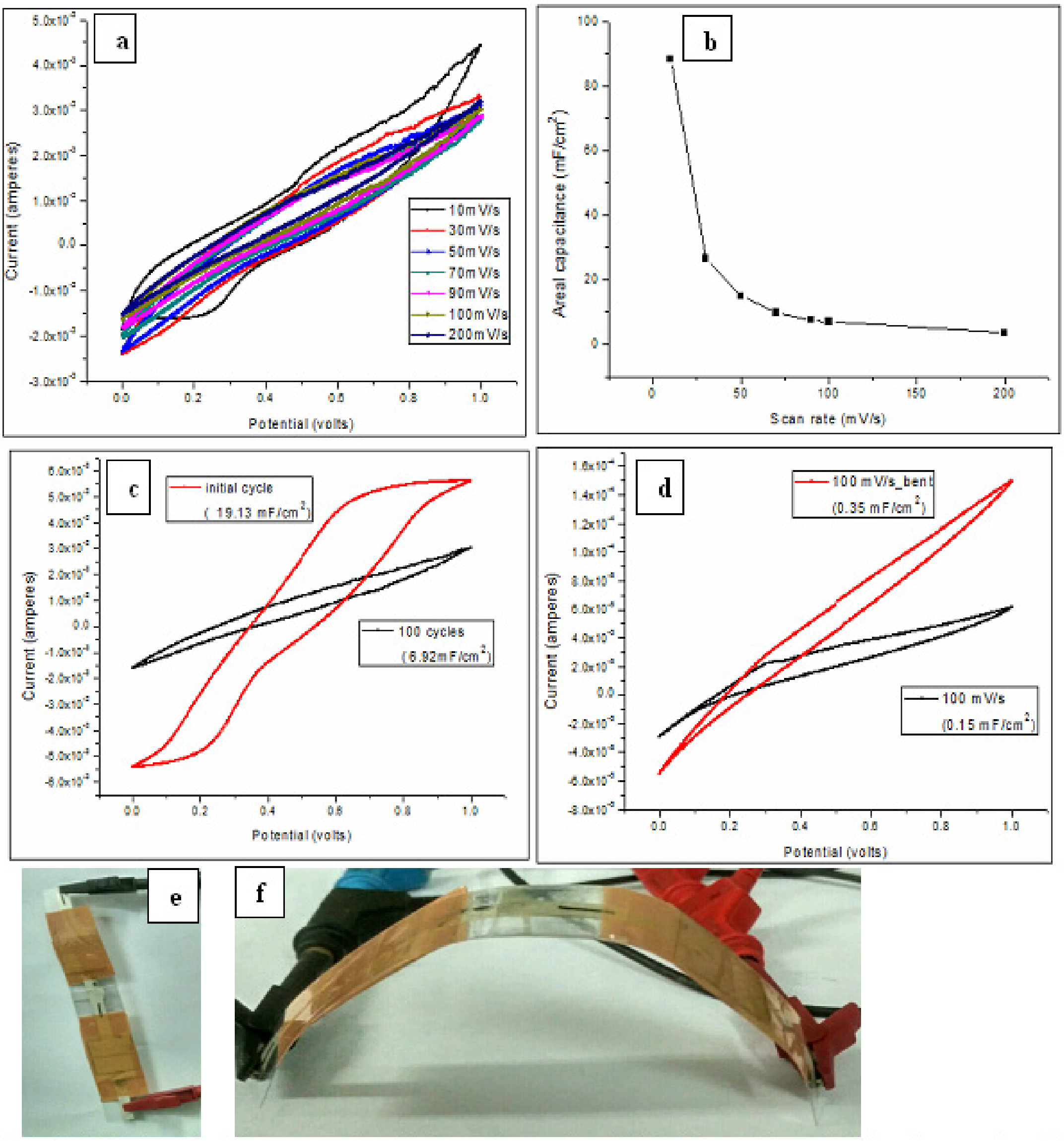

Figure 4. (a) CV profile of supercapacitor at different scan rates; (b) Areal capacitance at different scan rates; (c) Retention of capacitance after ~100 CV cycles at 100 mV/s; (d) Bending test of series-connected supercapacitor at 100 mV/s; Series-connected supercapacitor in (e) unbent and (f) bent condition.

4. Conclusions

A planar asymmetric supercapacitor was fabricated by incorporating few-layer graphene and activated carbon synthesized from cheaply available biomass (banana peel) along with transition metal oxides (NiO/Co_3O_4) as electrode materials. A low-cost screen printing technique was used to print the current collectors. Few-layer graphene has low resistance and therefore is considered to enhance charge distribution, improving performance of the device. Porous activated carbon provides a large surface area for charge storage, thereby enhancing capacitance. Our planar asymmetric supercapacitor demonstrated high areal capacitance (88.31 mF/cm^2 at 10 mV/s scan rate). The fabricated device demonstrated satisfactory performance under multiple cycling and bending conditions. Different combinations of electrode materials should be explored to get better cyclability and higher capacitance.

Acknowledgments

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References

1. H.I. Becker, Low voltage electrolytic capacitor. US Patent 2,800,616 (1957).
2. B.E. Conway, Transition from supercapacitor to battery behaviour in electrochemical energy storage. *J. Electrochem. Soc*. **138** (1991) 1539–1548.
3. X. Li & B. Wei, Supercapacitors based on nanostructured carbon. *Nano Energy* **2** (2013) 159–173.
4. J. Wang & S. Kaskel, KOH-activation of carbon-based materials for energy storage. *J. Mater. Chem.* **22** (2012) 23710–23725.
5. T. Purkait, G. Singh, M. Singh et al., Large area few-layer graphene with scalable preparation from waste biomass for high-performance supercapacitor. *Sci. Rep.* **7** (2017) 15239.
6. Y. Wang, J. Guo & T. Wang, Mesoporous transition metal oxides for supercapacitors. *Nanomaterials* **5** (2015) 1667–1689.
7. Chen, Q. Li, X. Zang et al., Effect of different gel electrolytes on graphene-based solid-state supercapacitors. *RSC Adv.* **4** (2014) 36253–36256.
8. B.D. Boruah, A. Maji & A. Misra, Flexible array of microsupercapacitor for additive energy storage performance over a large area. *ACS Appl. Mater. Interfaces* **10** (2018) 15864–15872.