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Knitted Graphene Supercapacitor and Pressure-Sensing Fabric †

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Abstract: This research utilizes a simple and effective dip coating/ultrasonication method to prepare porous graphene-coated sensing fabrics made with commercially produced acrylic/spandex yarn with multifunctional performance. We examine the electrochemical performance of graphene-coated fabrics and explore their potential in applications involving pressure sensors. The results show that our graphene-coated fabric demonstrates a maximum specific capacitance value of 17.4 F/g. When applied as a pressure sensor, the capacitance change rate of our sensor increases linearly with the increase in pressure applied to the fabrics. Our sensor also shows a fast response in a pressure loading–unloading test, which indicates an outstanding sensing property and shows promising capabilities as a supercapacitor.

Keywords: e-textiles; graphene; supercapacitor; pressure sensor

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1. Introduction

Stretchable and wearable electronics have attracted extensive attention from both academia and industry due to their unique flexibility, deformability, and portability [1]. In addition, development of energy storage devices with good electrochemical performance and flexibility to meet the energy requirements of flexible wearable electronics has become one of the major research directions in recent years due to the increasing prevalence of wearable technologies [2].

Among various conductive materials, graphene has become an ideal material for flexible electrodes because of their extremely high specific surface area, mechanical stiffness, and outstanding electrical conductivity [3,4]. The combination of textiles and graphene maintains the original physical properties of the fabric and provides a support for active materials while providing a stable conductive surface. Therefore, graphene-coated fabrics are considered an ideal material for electrodes [5].

In this study, we design and knit graphene-coated sensing fabrics with acrylic/spandex yarns through a cost-effective dip-coating method. The microstructure, and electrochemical and sensing properties of the porous graphene-coated sensing fabrics are examined to investigate their capabilities as a supercapacitor and pressure sensor.

2. Materials and Methods

2.1. Graphene-Coated Knitted Fabrics

We designed knitted fabrics with a full needle knitting structure, using a gauge 10 Dubied knitting machine, 25 rows in height, and with 45 needles on a double bed to knit the elastic yarn (90% acrylic and 10% spandex) into the same dimensions (20 mm × 60 mm × 1.5 mm).

The prepared knitted fabrics then were then fabricated into sensing fabrics using the dip-coating method, shown in Figure 1. The fabrics were purified by soaking them in ethanol and deionized water washings. After drying, the fabrics were dipped into a stable 1.2 wt.% graphene/acetone suspension under sonication for 30 min and then dried at room temperature. After three cycles of the coating process, the fabrics were washed with deionized water and then dried in a furnace at 40 °C.

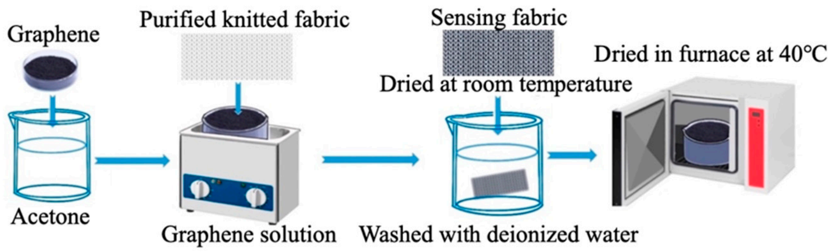

Figure 1. Fabrication process of graphene-coated sensing fabrics.

2.2. Electrochemical and Sensing Property Tests

The electrochemical properties of our graphene-coated fabrics were examined by testing cyclic voltammetry (*CV*), galvanostatic charge/discharge (*GCD*), cyclic stability measurements, and electrochemical impedance spectroscopy (*EIS*). A three-electrode supercapacitor device was set up, as illustrated in Figure 2, using 1 M Na2SO4 solution as the electrolyte, a Pt sheet as counter electrode, a Ag/AgCl sheet as reference electrode and our graphene-coated fabric as the working electrode. The *CV* curves were generated at a scan rate from 20 mV/s to 100 mV/s. The specific capacitance (*C*) was calculated from the area in the *CV* curves using the following equation:

$$C = \frac{\int I\, dv}{v \triangle Vm} \tag{1}$$

Using the same three-electrode setup, the *GCD* measurements were tested at current densities of 0.5, 1, 1.5 and 2 A/g. The specific capacitance value with the current densities in the *GCD* curves was calculated using the following equation:

$$C = \frac{I \cdot \triangle t}{m \cdot \triangle v} \tag{2}$$

where I = current (A), v = scan rate (V·s−1), V = working potential, m = weight of graphene in the fabrics (g), and Δt = discharging time.

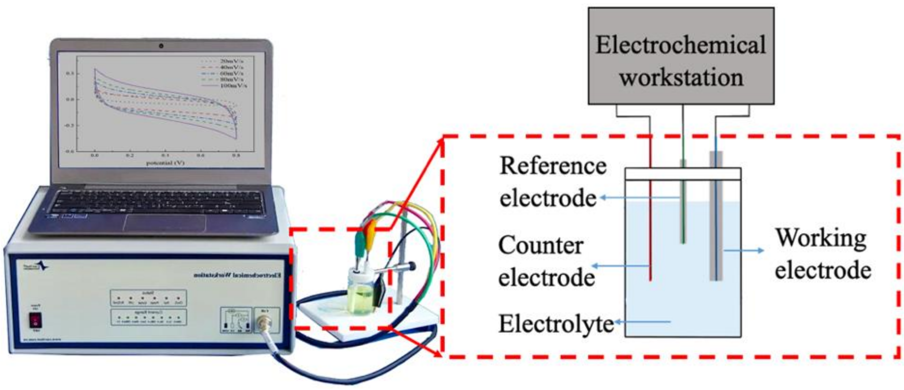

Figure 2. Schematic diagram of three-electrode supercapacitor system connected to electrochemical workstation.

The cyclic stability test was conducted at a current density of 2 A/g, charging and discharging 10,000 times.

The Nyquist curve was produced using the *EIS* and it measured the impedance through a decreasing AC frequency. The ion diffusion resistance of our graphene-coated fabric was calculated using Zview software.

For the sensing property test, various pressures were applied to the sensing fabric, and the electrochemical station was applied to measure the capacitance of the fabric. Then, the response time of our graphene-coated fabric was tested under external loading and unloading with a pressure of 24.5 kpa. The change rate in the capacitance was calculated using the ratio of change in capacitance ($\triangle C$) to the original capacitance (C_0).

3. Results and Discussion

3.1. Morphology of Graphene-Coated Fabrics

A scanning electron microscope (SEM) was applied to analyze the surface of our graphene-coated fabrics at different magnifications with the operating voltage of 5 kV.

Figure 3a is an overview of the knitted fabrics, showing porous structures that allows deposition of charged ions. It can be seen from Figure 3b that acrylic fibers form a uniform interweaved structure with multiple graphene nanoplatelets (GNPs) dispersed on the fiber surface and between fiber gaps. Figure 3c shows a typical crinkled structure of GNPs coating the textile fibers. Overall, the porous structure of our knitted fabrics can provide sufficient surface are for energy storage, which indicates the potential for incorporating such materials into a supercapacitor.

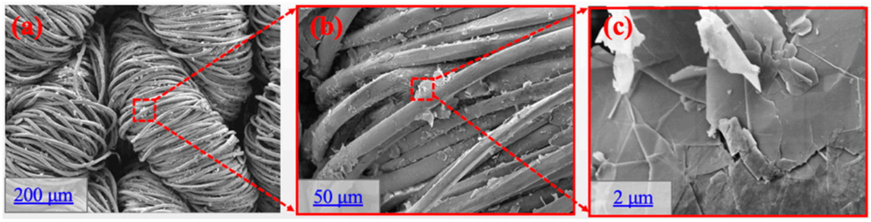

Figure 3. SEM images of graphene-coated knitted fabrics at (**a**) 200 µm, (**b**) 50 µm and (**c**) 2 µm.

3.2. Electrochemical and Sensing Performance of Graphene-Coated Fabrics

In the electrochemical performance tests, a *CV* test was first conducted, as shown in Figure 4a. All the *CV* curves at different scanning rates show a similar rectangular shape and demonstrate a maximum specific capacitance of 17.4 F/g at a scan rate of 20 mv/s. Figure 4b shows the *GCD* curves of graphene-coated fabrics. When the current density increases from 0.5 A/g to 2 A/g, the corresponding specific capacitance decrease from 16.7 F/g to 7.5 F/g. The *GCD* curves at different current densities all show similar trends with a triangular curve, indicating ideal capacitor performance. Figure 4c reflects the cyclic stability of the graphene-coated fabrics. After 10,000 cycles of charging and discharging, the capacitance of our sensing fabric remained above 90%. Figure 4d demonstrates the Nyquist curve of the graphene-coated fabrics. The almost negligible semicircle diameter for the graphene-coated fabrics reveals that the porous structure of the knitted fabrics facilitates a good contact between the electrode and electrolyte. The ion diffusion resistance of the graphene-coated fabric was calculated to be 20 Ω, demonstrating a good ion transport ability.

We further applied our graphene-coated fabric as a pressure sensor to test its sensing performance. Figure 5a shows that the capacitance change rate of our sensor increases linearly with the increase of pressure applied to the fabric. When the pressure was increased to 24.5 kPa, the capacitance change rate of the graphene-coated fabric reached its maximum value of 28%. Figure 5b demonstrates that our sensor had a fast response time with a recovery time of 0.6 s and 0.4 s under external loading and unloading, respectively. Our graphene-coated fabrics showed satisfactory performance as supercapacitors and as flexible sensors, indicating their great potential in further practical applications.

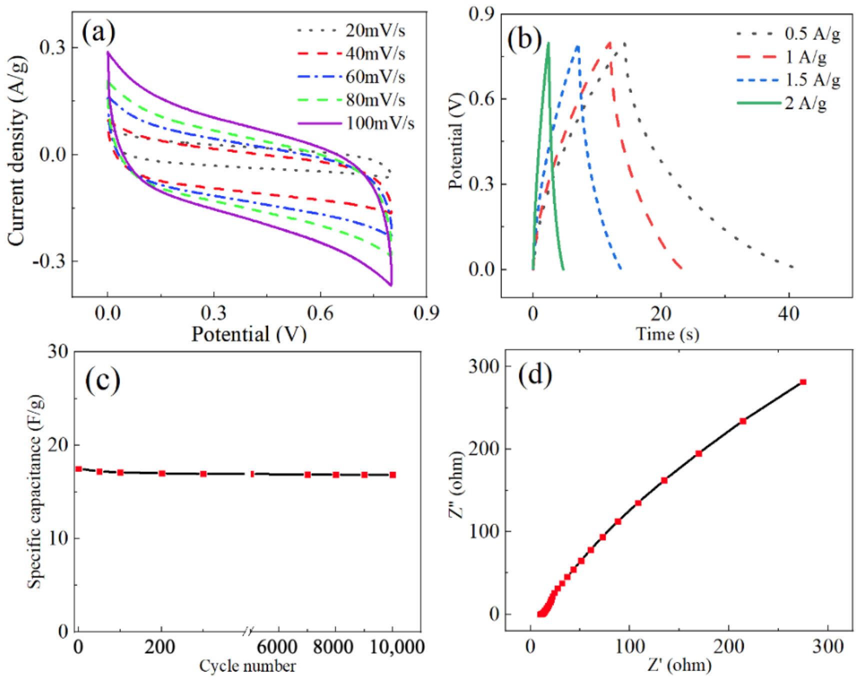

Figure 4. (**a**) CV curves of graphene-coated knitted fabrics at different scanning rates; (**b**) GCD curves of graphene-coated knitted fabrics; (**c**) cyclic stability test of graphene-coated knitted fabrics; (**d**) Nyquist diagram of graphene-coated knitted fabrics.

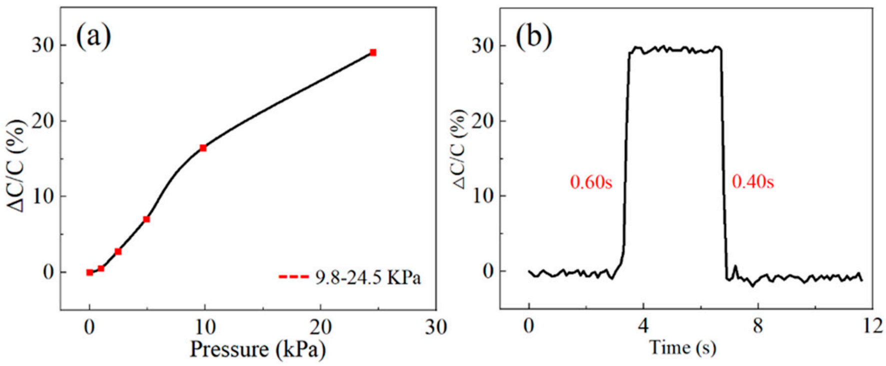

Figure 5. (**a**) Rate of change in capacitance as pressure applied within 0–25 kPa; (**b**) response and recovery time test under loading and unloading pressure.

4. Conclusions

This paper presents a graphene-coated sensing fabric that can be applied as a supercapacitor or pressure sensor by using a simple dip coating and ultrasonication method. The sensing fabric based on double bed weft-knitted fabric using acrylic/spandex yarns exhibits the ability to be charged/discharged. When applied as an electrode, our graphene-coated fabric demonstrates a maximum specific capacitance value of 17.4 F/g. Additionally, the graphene-coated fabric can be applied as a pressure sensor, the maximum capacitance change rate of our sensing fabric is up to 28%, and the compression response and recovery time are 0.6 s and 0.4 s. Since this research only explored the potential for graphene-coated fabric being applied as a supercapacitor or pressure sensor, further work will consider the stability and effect of wear due to washing. In addition, we will explore the reliability and longevity of our sensing fabric in a device or product.

Author Contributions: Conceptualization, Y.Z. and C.Z.; methodology, C.Z.; formal analysis, Y.Z.; investigation, Y.Z.; resources, Y.Z. and C.Z.; writing—original draft preparation, Y.Z.; writing—review and editing, R.S.; visualization, Y.Z.; supervision, C.M. and R.S.; project administration, C.M. and R.S. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest.

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