Preparation and Application of Flexible Conductive Fabric Based on Silk

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Abstract. Applications in wearable and implantable electronic information products have increased in recent years, and traditional conductive materials have poor flexibility. To meet the trend of flexible, wearable and implantable electronic devices, flexible conductive materials are widely accepted. Here, we report on a method of preparing a flexible conductive textile material. The graphene is finished onto the surface of the silk fabric by multiple impregnation-reduction methods, and the silk fabric retains good flexibility while obtaining conductivity. Flexible conductive silk also has a good temperature and strain response, and its electrical resistance changes with temperature and strain. The obtained flexible conductive silk fabric has the advantages of both silk fabric and graphene, it has broad application prospects in wearable devices, health care monitoring, and human-machine interface.

1. Introduction

With the development of science and technology, textiles have broken through the category of warmth and beauty. Textile products are no longer limited to use as clothing or home decoration. With the development of high-performance textile products in recent years, textile fabrics have expanded from traditional fields to military, civil aviation and other fields. Conductive finishing of textile products to obtain excellent electrical conductivity can not only effectively expand the application range of textile materials, but also meet the development trend of today's flexible electronic materials, and provide an ideal flexible conductive material for the development of flexible electronic equipment [1-2].

In 2004, graphene was successfully prepared in the form of monolayer by mechanical stripping method and attracted attention in various fields of scientific research [3-4]. The good flexibility and mechanical properties of graphene and its excellent electrical conductivity make it the best choice for the preparation of flexible conductive materials. In the actual research process, the preparation of graphene conductive materials is usually based on selected flexible materials [5-8]. Graphene, GO or RGO is a conductive medium, and the conductive medium is composited with the substrate by a suitable finishing process, and then subjected to subsequent treatment to obtain a flexible conductive material to be prepared [9-11].

Silk is a natural protein fiber that has been used as a textile for more than 4,000 years. Silk not only has the general properties of traditional textile fibers but also has excellent biocompatibility and
biodegradability. This unique advantage has made silk popular in the study of wearable and implantable electronic information products [12-14].

In this paper, we have finished the graphene onto the surface of the silk fabric by multiple impregnation-reduction methods to obtain the conductivity of the silk fabric.

2. Experimental section

Materials: Graphite powder (Alfa Aesar Co., Ltd.), concentrated sulfuric acid (95%–98%), potassium permanganate (National Pharmaceutical Group Chemical Reagent Co., Ltd.), sodium nitrate (National Pharmaceutical Group Chemical Reagent Co., Ltd.), hydrogen peroxide (30 %, Sinopharm Chemical Reagent Co., Ltd.), hydrochloric acid (36%–38%, Sinopharm Chemical Reagent Co., Ltd.), stannous chloride (National Pharmaceutical Group Chemical Reagent Co., Ltd.).

2.1. Preparation of graphene oxide

In this experiment, the graphene was prepared by the Hummers method. The preparation of GO was divided into three steps, as shown in Figure 1:

In the first step, 1 g of graphite powder, 23 ml of concentrated sulfuric acid and 0.5 g of sodium nitrate were taken, poured into a 250 ml three-necked flask, placed in an ice water bath, stirred at 4 °C for less than half an hour, and then added to the high manganese in portions during the stirring. Potassium acid 3g, after the addition was completed, the reaction was kept for 2 hours to complete the low-temperature reaction.

In the second step, the temperature of the system was raised to 35 °C, and the reaction was kept for 3 hours. Complete the medium temperature reaction.

In the third step, the temperature of the system was raised to 98 °C, and 46 ml of distilled water was added dropwise with a constant pressure funnel, and the reaction was carried out for 1 hour to complete the high-temperature reaction.

After the reaction, 70 ml of distilled water and 10 ml of hydrogen peroxide were added, stirring was continued for 15 minutes, followed by hot filtration, and the product was washed with a large amount of dilute hydrochloric acid (5% by volume) and distilled water until the pH was close to 7. Finally, the obtained product was centrifuged. After washing, it was placed in a drying oven, dried at 60° for 24 hours, and taken out to obtain GO powder.

![Figure 1. Preparation process of graphene oxide](image)
2.2. Preparation of flexible conductive silk
Preparation of graphene oxide dispersion: a certain amount of graphene oxide is dissolved in water, and
a certain concentration of graphene oxide dispersion is prepared and placed in an ultrasonic machine for
half an hour.

Dipping process: The silk (6 cm × 6 cm) fabric was put into the fabric, immersed for 1 hour at room
temperature and stirred every 5 minutes. After the end, the silk was taken out and dried under
vacuum at 60 °C for 30 min.

Reduction process: stannous chloride 1.6mol/L, 0.5ml of reducing agent is added dropwise to GO
finishing dried silk fabric by drop coating method, and dried in a drying oven at 60 °C for 30 minutes,
the whole reduction process is maintained. In contact with air. Finally, the by-product was washed away
with ethanol and dried in an oven at 60 °C for 30 minutes to obtain a conductive silk fabric.

We successfully prepared graphene oxide by the improved Hummers method. Raman spectroscopy
of self-made graphene oxide (Fig. 2a), D-peak and G-peak are the Raman characteristic peaks of C atom
crystals, D-peaks represent the defects of C atomic lattice, and G-peaks represent the in-plane stretching
vibration of C atom sp2 hybridization, the G peak of GO is around 1586cm-1, which reflects the
symmetry of the graphite structure, and the D-peak is near 1348cm-1, which reflects the disorder of the
inner layer of GO; ID The larger the value of IG=1.04, the more damage the C=C bond of C atom crystal,
the more defects, indicating that the graphite layer is intercalated with more abundant oxygen-containing
functional groups during oxidation. =C causes damage and forms a defect. The XPS full spectrum of
graphene oxide (Fig. 2b) shows that C and O are the main elements.

3. Results and discussion
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![Figure 2. (a) Raman spectroscopy of GO. (b) XPS spectra of GO](image-url)

After drying the graphene-impregnated silk fabric, the color becomes yellow, and after the stannous
chloride solution is reduced, the surface of the silk cloth is uniformly black, and the color gradually
depens as the number of times of the immersion-reduction increases (Fig. 3c). When the number of
immersion-reduction times is less than seven, the conductivity of the silk fabric is poor, and the
conductivity is remarkably increased after the immersion-reduction seven times (Fig. 3b). This is
because when the number of immersion-reduction times is low, the amount of RGO deposited on the
surface of the silk is small, the regularity and continuity of RGO on the surface of the silk are poor, and there are some cracks between the RGO sheet and the sheet, so the conductivity is poor. After the immersion-reduction times are more than seven times, the conductivity of the silk fabric increases slightly with the increase in the number of immersion-reduction times. From the viewpoint of simplifying the process, the number of immersion-reduction times is seven times. When the concentration of the graphene oxide solution is low, the obtained silk fabric has a large electric resistance, and the electric resistance gradually decreases as the concentration increases, and the graphene oxide solution is obtained by comparing the electric resistance of the silk fabric prepared by different concentrations of the graphene oxide solution. The optimum concentration is 3 wt%.

![Figure 3](image1.png)

**Figure 3.** (a) Effect of GO concentration on conductivity. (b) Effect of the number of immersion on conductivity. (c) A picture of the impregnated-reduced conductive silk fabric. (d) Flexible conductive silk fabric resistance test.

![Figure 4](image2.png)

**Figure 4.** (a) Effect of temperature on electrical resistance of flexible conductive silk fabrics. (b) Effect of strain on electrical resistance of flexible conductive silk fabrics

The electrical resistance of the flexible conductive silk fabric after the impregnation-reduction finishing varies with changes in temperature and strain. This is due to the thermal expansion and contraction of the graphene coating on the surface of the silk and is sensitive to temperature changes (Fig. 4a). When the strain of the silk fabric changes, the continuity of the graphene coating on the surface of the silk changes, which affects its electrical resistance (Fig. 4b). Due to the temperature and strain response of the flexible conductive silk fabric, the practicality of the finished flexible conductive silk fabric can be extended to sensors for touch and breathing.
4. Conclusion
Silk is one of the most abundant natural protein fibers, with many advantages such as lightweight, good flexibility and good biocompatibility. As a high-performance synthetic inorganic nanomaterial, graphene is known for its extraordinary thermal conductivity and electrical conductivity. We have successfully prepared better quality graphene oxide by the improved Hummers method. The graphene was finished onto the surface of the silk fabric by repeated impregnation-reduction methods to impart conductivity to the silk fabric. The optimum concentration of the graphene oxide solution was 3 wt%, and the optimum number of impregnation reductions was seven times. Flexible conductive silk also has a good temperature and strain response, and its electrical resistance changes with temperature and strain. The advantages of the obtained flexible conductive silk fabric combined with silk fabric and graphene show broad application prospects in wearable devices, health care monitoring, and human-machine interface.

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