ABSTRACT

To address the issues of poor graphene dispersion, uneven thermal conductivity, and the environmental effect of porous polyurethane (PU) solutions, natural nano cellulose is employed as a surfactant to dissolve the graphene slurry in order to build a composite heating film. By altering the volume of the graphene slurry, the screen-printing method performs in-situ coating on heat-reflective cloth (sportswear lining materials) and determines the heating impact and washing qualities of the clothing. The results reveal that natural nanocellulose has a good dispersion effect. After the addition of silver paste, graphene dispersions with varying concentrations exhibit good thermal and electrical conductivity. When the heating voltage is 8v and the graphene slurry concentration is 12.5 % (wwt), the surface temperature of the heating film can exceed 50°C while the power consumption is low, which not only maintains long-term power supply but also addresses the shortcomings of the traditional polyester heating film, such as uncomfortable wearing. Furthermore, even after washing and soaking it more than 50 times, it has an excellent heating function.

Keywords: Nano cellulose; Graphene; Thermal conductivity; Dispersion effect.
Introduction

Graphene is a two-dimensional carbon lattice that has attracted much attention due to its excellent mechanical, thermal, and electrical properties. Ideal graphene conductive paste can produce flexible electric heating elements, sensors, cooling elements, and many other popular products. In the field of information, intelligent clothing and clothing has an important role. Among them, metal paste, conductive polymer paste, and carbon composite paste are the most widely used [1]. Junlin Ma [2] et al. developed Prussian blue/graphene ink by grinding and assembling composite materials, which can be used for screen printing flexible biosensors and supercapacitors. Based on the rapid electron transfer of Prussian blue/graphene ink during electro-catalysis, the biosensor can effectively monitor \( \text{H}_2\text{O}_2 \) and acid substances with a long linear range, high sensitivity, and low detection limit. These biosensors can be used for detection in real life [3]. Hua uses a mixture of graphene and carbon black to make a composite conductive paste and sets up multiple sets of pastes with different proportions of graphene and carbon black. The results show that the graphene and carbon black composite conductive paste has good stability and can be stored for a long time [4]. The paste can be combined with any substrate through screen printing, but the dispersion of this graphene composite paste is not ideal [5].

In recent years, the key research of flexible electric heating elements focuses on the flexible electric heating elements that have electrical functions such as heating and conduction, but also can maintain the original physical properties of the fabric, such as tactile softness, maintain the original organizational structure, and have the functions that people often need, such as permeability, friction resistance, water-resistance and so on [6]. The relatively ideal method at present is the coating method, because it requires little to the base of the heating element, unlike conductive metal fibers, which require additional braiding. This method only needs to coat the slurry on the selected substrate so that the original physical properties of the fabric can be retained and the electrical function of the fabric can be achieved [7]. Inkjet printing, scraping, impregnation, and screen printing are some commonly used coating methods for flexible electric heating elements. Among existing mass printing technologies, screen printing is considered to be the most versatile and mature, simpler and faster than other printing tools [8]. Screen printing is the process of evenly filling the paste onto the edge surface of the mold, which is located on the desired substrate and held in place by a gasket. During the printing process, the scraper aligns
with the mold and pushes the paste through the patterned screen to produce the desired pattern, which is transferred to the substrate after a single pass [9]. Screenprinting has the advantages of simplicity, repeatability, and high compatibility with various inks and substrates: making it a cost-effective method for large-scale printing with flexible equipment [10].

Graphene has good conductivity, thermal conductivity, and toughness, so graphene is very suitable for conducting flexible electric heating elements. In this project, graphene and nanocellulose are combined to explore whether nanocellulose, a green environment-friendly material, can be used to replace toxic solvent and combine with graphene to make graphene conductive paste with good dispersion, which is not easy to aggregate and can be preserved for a long time [11]. The temperature changes of four graphene heating films with different concentrations under different voltages were used to explore the effects of different voltages, graphene concentration in graphene slurry, and resistance on the temperature of graphene heating films [12].

1. Experimental Part

1.1. Preparation of Graphene Slurry: To find out the most suitable to be added into clothing as clothing electric heating film, nanocellulose was used as a solvent (Cellulose Nanofibers or CNF) to prepare four kinds of graphene conductive paste with different graphene concentrations: 7.5%, 10.0%, 12.5%, and 15% respectively. Each of the four concentrations of slurry is equipped with 50g.

<table>
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1.2. Graphene Paste Coating: For printing, the screen (size of 15cm×10cm rectangular screen) is placed on the thermal reflective cloth, and the adjusted graphene paste is
poured evenly on one side of the screen. The heating and blowing function of the electric hairdryer is dried, and different layers of graphene are printed according to the design. The number of layers of graphene film should not be too much, too much will lead to the overall hard and brittle graphene film, with the movement of the base cloth graphene film is easy to crack.

1.3. Performance Test of Graphene Heated Film:

1.3.1. Film Resistance Test: To control and debug the resistance on the surface of the film, to maintain high heat, low energy consumption, and continuous heating, this experiment uses a regulated DC power supply to supply power to the heating film, and a multimeter to measure its surface resistance.

1.3.2. Heating Temperature and Infrared Imaging Test: The heating of the film was tested by an infrared thermal imager. Four prepared heating films with different concentrations were heated by the power supply. An infrared thermal imager was used to select the film with the most uniform heating among the four graphene heating films with different concentrations as the test object of this experiment. In the experiment, a complete graphene film was evenly divided into four equal areas, and 5-10V voltages were applied to different concentrations of graphene. Temperature data of the four equal areas were measured, and the average temperature of the four areas was taken as the result of temperature measurement.

1.4. Wash-ability Analysis and Test: Nano-coating was added in water, and the surface of graphene heated film prepared was coated by the impregnation method and then dried. A wear resistance test was carried out using a testing machine, and its heating was measured after 50 times.

2. Conclusion and Discussion:

2.1. The Heating Rate of Graphene Heating Film: According to the voltage and power range provided by the charging bank used in the current heating clothing market in India, a 5-10V power supply is mainly used for testing in this study. The heating situation of four graphene heating films with different concentrations is shown in Figures 3 to 6. Due to the rapid temperature rise of graphene heating film, the temperature rise of graphene
heating film with different concentrations was tested by an infrared thermal imager every 30 seconds when different voltages were applied. The experiment was carried out at room temperature of 25°C. After each test, the temperature of the membrane was restored to room temperature before the next test (repeated three times in total).

**Figure 1.** 7.5% Graphene heated film changes

**Figure 2.** 10% Graphene heated film changes
Figure 3. 12.5% Graphene heated film changes

Figure 4. 15% Graphene heated film changes
It can be seen from the data in Figures 1 to 4 that, at the same concentration, the heat released by the heating film increases with the increase of the heating voltage. The temperature of the graphene heating film with a concentration of 7.5% is low at 5V, only 27°C. With the increase of the voltage, the temperature can reach 33.3°C when the voltage reaches 10V. In addition, the temperature rises rapidly, approaching the maximum value in the 60s, and then tends to be stable. The temperature of the graphene-heated film with a concentration of 10% can reach 30.7°C at 5V. With the increasing voltage, the temperature can reach 46.2°C when the voltage reaches 10V, and the temperature increases rapidly, and gradually becomes stable after approaching the maximum value in the 30s. The temperature of the graphene heated film with a concentration of 12.5% reaches over 30°C when the voltage is 5V and 67.3°C when the voltage reaches 10V with increasing voltage. The temperature of 15% graphene heated film reaches 39.6°C at 5V and 72°C at 10V with increasing voltage.

2.2. Relationship between Graphene Heating Film Temperature and Graphene Concentration: According to the experimental data in Figures 5 to 8, the temperature of the graphene heating film is related to the concentration of graphene. At the same voltage, the higher the concentration of graphene, the higher the temperature of the membrane. Among the four concentrations of 7.5%, 10%, 12.5% and 15%, the temperature of the 15% concentration of graphene film are higher than that of the other concentrations of graphene film at various voltages, and the temperature of 7.5% concentration of graphene film is the lowest. The higher the voltage is, the greater the temperature difference of different concentrations of graphene film is. At 5V, 15% film temperature is 12.6°C higher than 7.5% film temperature. At 6V, the film temperature of 15% was 16.4°C higher than that of 7.5%. At 7V, 15% film temperature is 23.6°C higher than 7.5% film temperature. At 8V, 15% film temperature is 26.1°C higher than 7.5% film temperature. At 9V, the film temperature of 15% was 35.9°C higher than 7.5%. At 10V, 15% film temperature is 38.7°C higher than 7.5% of the film.
Figure 5. Graphene heated film changes in 5V

Figure 6. Graphene heated film change in 6V
Figure 7. Graphene heated film change in 7V

Figure 8. Graphene heated film change in 8V
3. **Summary:** In this research, four different concentrations of graphene-heated film products of 7.5%, 10%, 12.5%, and 15% were investigated by mixing graphene powder with nanocellulose. To investigate the effects of different voltages, graphene concentration in graphene slurry and resistance on the temperature of graphene heating films, the temperature variations of four graphene heating films with varied concentrations were observed under different voltages. The following conclusions may be taken from the experiment and examination of the experimental data:

i. The combination of graphene and nanocellulose can produce graphene slurry with good disparity and difficulty in aggregation.

ii. In situ screen printing allows the paste to be coated on a soft substrate, a thermal reflective cloth, to produce the finished graphene heating film. The temperature of the graphene heating film is related to the concentration of graphene in the graphene slurry. The higher the concentration of graphene in the graphene slurry is, the higher the temperature of the graphene film will be when other conditions remain unchanged. For example, at 10V, the temperature of 15% graphene heating film is 38.7°C higher than that of 7.5% graphene heating film.

The temperature of the graphene heating film is related to the applied voltage. The higher the applied voltage is, the higher the temperature of the graphene film will be when other conditions remain unchanged. The temperature of 15% graphene heated film is 72°C at 10V and 39.6°C at 5V. The temperature at 10V is 32.4°C higher than that at 5V.

**Conflict Of Interest**

The authors have no conflict of interest.

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References


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