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Thermal transfer printing of laser-induced graphene for textile supercapacitors

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In this work, thermal transfer printing process was performed to fabricate a LIG on textile substrates (LIGT). A thermally adhesive film of a conventional transfer paper was used to attach a LIG on polymer substrates (LIGP) to a cotton fabric substrate at elevated temperature and pressure of thermal transfer printing. Experimental conditions were optimized to provide LIGTs with low sheet resistance while maintaining high porosity and crystallinity. As-fabricated LIGTs were demonstrated as in-plane textile MSC electrodes by exhibiting typical electrical double layer capacitive characteristics. The proposed thermal transfer printing is also scalable to make large-area LIGT patterns, thus providing various array configurations of LIGT-MSCs in series and in parallel for high voltage and large capacitance textile MSCs, respectively.

1. Introduction

Electronic textiles have garnered extensive attention in various applications such as sensors, actuators and energy storage devices [1-3]. In order to realize electronic textiles, textile energy storage devices as power sources need to be fabricated and integrated with other active textile devices. Microsupercapacitors (MSCs), thin-film energy storage devices, have received a great deal of attention as power sources for wearable, stretchable or textile energy storage devices due to their fast charging capability, long life cycle, and good safety.

Laser-induced graphene (LIG), obtained by high power laser irradiation on a conventional polyimide (PI) film, have been widely researched as electrode materials of flexible MSCs [4-5]. Various carbon precursors such as polymer films, cloth, bread or wood were carbonized by direct laser writing to form conductive LIGs [6]. Moreover, LIGs were transferred on stretchable rubber substrates to provide stretchable electrodes [7]. However, few efforts on LIG-based textile MSCs (LIG-MSCs) have been reported.

In this work, thermal transfer printing process was performed to fabricate a LIG on textile substrates (LIGT). A thermally adhesive film of a conventional transfer paper was used to attach a LIG on polymer substrates (LIGP) to a cotton fabric substrate at elevated temperature and pressure of thermal transfer printing. Experimental conditions were optimized to provide LIGTs with low sheet resistance while maintaining high porosity and crystallinity. As-fabricated LIGTs were demonstrated as in-plane textile MSC electrodes by exhibiting typical electrical double layer capacitive characteristics. The proposed thermal transfer printing is also scalable to make large-area LIGT patterns, thus providing various array configurations of LIGT-MSCs in series and in parallel for high voltage and large capacitance textile MSCs, respectively.

2. Experimental details

2.1 Fabrication of LIG on a textile substrate

To fabricate LIG on a textile (LIGT) substrate, high power carbon dioxide (CO_2) laser was first irradiated on a commercial PI film to form a LIG on a polymer film (LIGP). CO_2 laser patterning produced high quality graphene-like carbon electrode with high crystallinity, low defect and low sheet resistance of 60 Ohm/sq.

A LIGT was fabricated by two successive thermal transfer printing process shown in figure 1. Thermal transfer printing process, known as T-shirt printing, employed material transfer by difference in adhesion forces under elevated temperature and pressure condition. In the proposed process, an adhesive layer of a transfer paper was first formed onto the textile substrate by removing a cover paper after the first thermal transfer process. The second thermal transfer process is performed on the adhesive layer area by using the previously prepared LIG pattern. By detaching the PI film, LIG pattern on the PI film is successfully transferred onto the textile substrate.

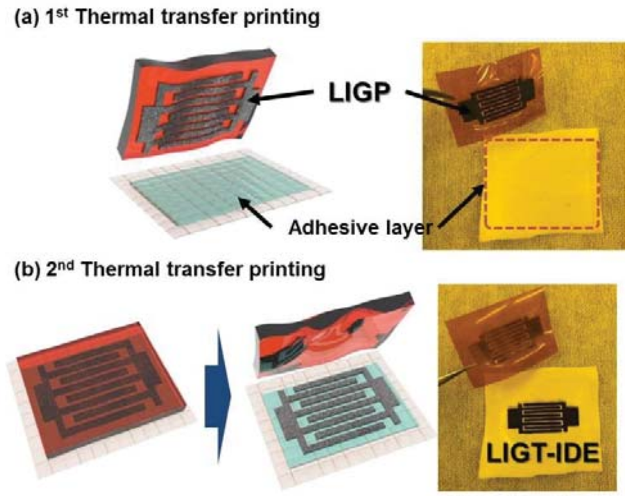

Fig. 1 Process sequence of thermal transfer printing for fabrication of LIGT electrode

2.2 Characterization of LIGT

Figure 2(a-f) shows the scanning electron microscope (SEM) images of LIGP and LIGT. Porous structure of LIG was maintained after thermal transfer process. Applied pressure was optimized to show the lowest sheet resistance of the LIGT (see fig 2(g)).

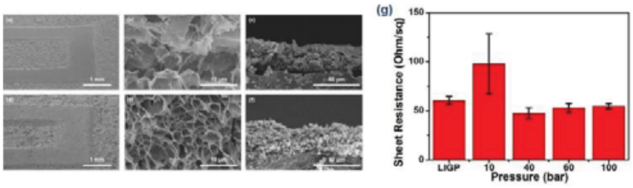

Fig.2 SEM images of (a-c) LIGP and (d-f) LIGP. (g) Sheet resistance of LIGT according to the applied pressure

2.3 Electrochemical performance of LIGT-MSCs

A LIG-MSC was fabricated using the LIGTs as two electrodes and a gel electrolyte of polyvinyl alcohol (PVA) in phosphoric acid (H3PO4) as an electrolyte, respectively. Representative electrode length (L), width (W_e), gap between electrodes (W_g), and number of finger electrodes (N) of a LIGT-MSC were selected as 2 mm, 1.5 mm, 0.5 mm, and 8, respectively.

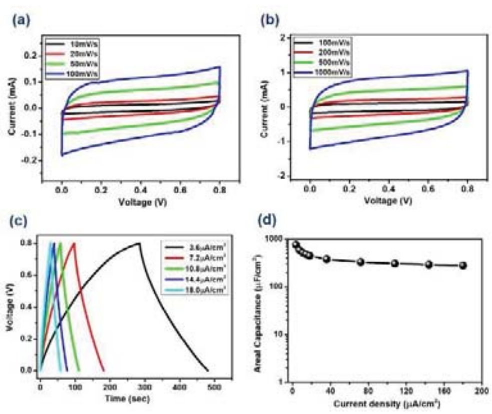

Fig.3 Electrochemical performance testing of LIGT-MSCs. (a-b) Cyclic Voltammetry (CV) and (c-d) Charge-Discharge (CD) testing

Figure 3 shows the results of electrochemical performance tests of a LIGT-MSC. Quasi-rectangular CV curve shapes were observed in a wide range of scan rates (10 mVs^{-1} ~ 1000 mVs^{-1}), indicating a typical electric double layer capacitor characteristic of the LIGT-MSC (Fig. 3a,b). Galvanostatic Charge-Discharge (CD) test results at various current densities (3 ~ 180 µA cm-2) also demonstrated that the LIGT-MSCs exhibited electrochemically capacitive behavior with quasi-triangular CD curves (Fig. 3c, d). The C_A from the CD curve was measured to be 0.76 mF cm^{-2} and 0.28 mF cm^{-2} at current densities of 3.6 µA cm^{-2} and 180 µA cm^{-2}, respectively. The obtained C_A (0.5~0.76 $mFcm^{-2}$) is comparable to that of other graphene-based MSCs [36-40]. Furthermore, bending cyclability of a LIGT-MSC exhibited capacitance retention of 96% after 1,000 cycles of large bending deformation (180 degree) shown in fig 4.

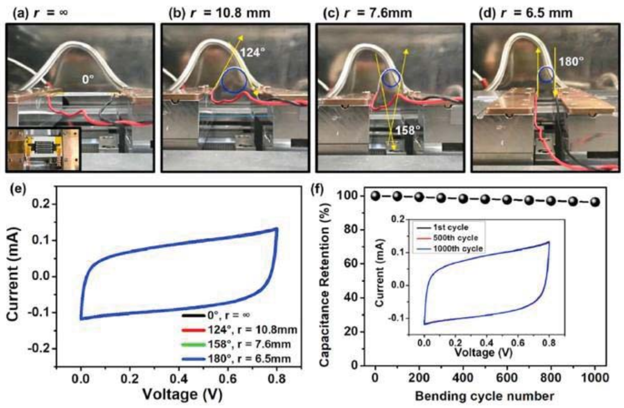

Fig.4 (a-d) Photographs of LIGT-MSC in different bent states. (e) CV curves of LIGT-MSC with different bending angle and radius. (f) Capacitance retention of LIGT-MSC during 1000 bending cycles compared to the value in flat state.

To extend LIGT-MSCs into practical applications as textile energy storage devices, the voltage and capacitance of the LIGT-MSCs need to be controlled depending on the application. An array of LIGT-MSCs with different configuration in parallel and series was fabricated by the proposed thermal transfer process. Figure 5 shows the different configuration of LIGT-MSCs and their electrochemical performances. The results exhibited that LIGT-MSCs could be configured to control the voltage and capacitance by connecting multiple LIGT-MSCs in series and parallel, respectively.

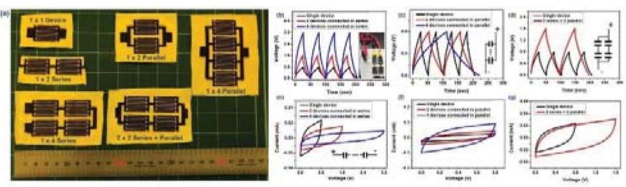

Fig.5 (a) Photographs and (b-g) electrochemical performances of LIGT-MSCs with different LIGT electrode array configurations.

3. Conclusions

In this study, a thermal transfer process was proposed as an inkless fabrication of textile MSCs. By using thermal transfer process, LIG formed on a polymer substrate was successfully transferred to the textile substrate. The LIGT preserved porous structure with 3D interconnected pores of LIGP with high crystallinity and low defects.
A LIGT-MSC showed areal specific capacitance of 0.76 mF cm^{-2} and capacitance retention of 96% after 1,000 cycles of large bending. Furthermore, LIGT-MSCs demonstrated the possibility of practical usage such as cyclic stability, and control of the working voltage and capacitance

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