Highly conductive and ultra-durable electronic textiles via covalent immobilization of carbon nanomaterials on cotton fabric

Junshuo Cui and Shuxue Zhou*

Graphene and multi-walled carbon nanotubes (MWCNTs) are promising candidates for the fabrication of flexible electronic textiles (E-textiles) due to their low weight and chemical stability. However, common issues of the carbon-based E-textiles such as low electrical conductivity, poor durability, and tedious fabrication processes still exist at present. In this study, fabrics with low sheet resistance and ultra-durability were fabricated using a simple dip-coating approach. A self-cross-linkable dispersant, possessing several silanol groups, was used for preparing graphene and MWCNTs dispersions. After a dip-coating/drying process, covalent networks were formed across the coating layer, which firmly immobilized graphene and MWCNTs on the fabric surface. Conductive fabrics with a sheet resistance of 33.2 \( \Omega \, \text{sq}^{-1} \) and 29.8 \( \Omega \, \text{sq}^{-1} \) were obtained using MWCNTs and graphene as carbon nanomaterials, respectively. Their conductivities remained almost unchanged after washing and sonication in water and various organic solvents. The tensile strength of the fabric significantly improved after the incorporation of graphene or MWCNTs on its surface. Excellent stretch sensing and electric heating performances were also revealed for the nanocarbon-coated textiles.

Introduction

E-textiles are an intriguing research interest due to their potential applications in wearable electronics, flexible circuits, precision healthcare, power generation, and energy storage. For the fabrication of universal E-textiles, the conductive component is the most important factor that should be taken into consideration. Thus, metals, conductive polymers, and carbon materials are now commonly used. Metals such as silver and copper that possess excellent electrical conductivity are suitable for preparing highly conductive textiles. However, their low chemical stability and high cost are significant issues. Conductive polymers also exhibit some limitations because their conductivity may be lost under certain conditions. For example, the conductivity of the widely used poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PSS) severely deteriorates at a relative high temperature (120 °C) and polyaniline is less conductive in alkaline media due to undoping. In contrast, carbon materials, due to their good electrical conductivity, chemical stability, light weight, and cost efficiency, have been widely adopted for E-textiles.

There are three general approaches for the fabrication of carbon-based E-textiles: (1) weaving or knitting carbon fibers and yarns into full textiles, (2) carbonizing ready-made fabrics at high temperature under an inert atmosphere, and (3) coating readily available fabrics with carbon nanomaterials. Among them, the last approach has distinct advantages because nearly all fabrics can serve as the substrate without an additional weaving or knitting process, and conductive textiles can preserve their flexibility and mechanical strength, avoiding carbonization-induced brittleness. In addition, this approach can be achieved using various coating techniques, including drop-casting, brush-coating, blade-coating, dip-coating, spray-coating, screen-printing, and inkjet-printing. Furthermore, complex conductive images or logos can be finely created on textile surfaces. Overall, coating with carbon nanomaterials is convenient for the fabrication of E-textiles, which fulfills most of the requirements for universal utilization.

Graphene and CNTs (nanocarbons) are two types of attractive carbon nanomaterials, which are highly conductive and widely used for the fabrication of E-textiles through the coating approach. A crucial factor that determines the final quality of conductive fabrics is the uniform coating of nanocarbons. This further depends on the dispersion state of the nanocarbons and their interaction with the fabric. Homogeneous dispersion and strong interaction can result in high-quality E-textiles. Thus, numerous graphene-based E-textiles have been prepared using...
graphene oxide (GO) as the conductive precursor because the presence of abundant hydrophilic groups in GO not only facilitates the formation of a uniform dispersion but also results in interaction with the fabrics. However, the time-consuming reduction process for GO is indispensable to recover its conductivity. Besides, toxic reagents such as hydrazine and hydriodic acid are often used in the chemical reduction process. Consequently, possible residuals may be harmful to skin when the E-textiles contact with the human body. Also, the thermal reduction of GO often requires a relatively high temperature (about 200 °C), which inevitably damages the mechanical strength of the final conductive fabrics. Although the GO route is currently popular to obtain graphene-based E-textiles, it is actually not always the optimal choice.

Another option for the coating route is the direct utilization of nanocarbon dispersions, which is supposed to be the most convenient approach. For example, based on a PSS-stabilized reduced GO (rGO) dispersion, the scalable production of E-textiles was realized via a dip-coating and pad-dry technique. However, due to the existence of strong π-π interactions and van der Waals forces, it is difficult to obtain homogeneous graphene dispersions at high concentration. Thus, only a few graphene-based E-textiles have been fabricated via this approach. In contrast, several conductive fabrics have been prepared using CNT dispersions although they suffer the same problem. To obtain homogeneous CNTs dispersions, functionalization of pristine CNTs is frequently adopted. Surfactants or specific polymers are also widely adopted as stabilizers. Nevertheless, besides the cost for obtaining nanocarbons dispersions, two basic issues need to be addressed.

First, it is difficult to obtain a nanocarbon-based E-textile with a very low sheet resistance, particularly that based on graphene. For example, the sheet resistance of the rGO-based E-textile produced by Karim et al. was as high as 36.94 kΩ sq⁻¹ after five padding passes. In another case with the GO route reported by Lu et al., the sheet resistance of the E-textile was 1.5 kΩ sq⁻¹ after five coating-reduction processes. These sheet resistances are too high to meet the standard of some specific applications. Second, the durability of E-textiles is another big concern. Due to the lack of strong interactions between nanocarbons and fabric surfaces, pretreating the fabric or use of an intermediate reagent are often adopted to enhance the coating fastness. Although several studies claimed that the washing-durability of E-textiles have greatly improved due to the existence of hydrogen bonding or electrostatic attraction, their sheet resistance still remarkably increased after several washing tests.

Thus, herein, to obtain E-textiles with high conductivity and ultra-durability, we immobilized graphene and MWCNTs on a cotton fabric surface through covalent bonding. Self-cross-linkable nanocarbon dispersions with a high concentration were first prepared using a TSiPD⁺ dispersant, which contains abundant silanol groups (Fig. 1). By dip-coating cotton fabrics in the dispersions, sheet resistances as low as 33.2 Ω sq⁻¹ and 29.8 Ω sq⁻¹ were obtained for the MWCNTs- and graphene-based E-textiles, respectively. In addition, the dehydration condensation of the silanol groups caused the nanocarbons to be firmly bound on the fabric surfaces. The sheet resistance of the textiles remained nearly unchanged after long-time washing and sonication test, exhibiting excellent durability.

**Experimental section**

**Materials**

(3-Glycidoxypropyl)trimethoxysilane (GPTMS, 97%), p-phenylenediamine (PDA, 99%), anhydrous methanol, formic acid (88%), sodium dodecyl sulfate (SDS, 99%), polyvinylpyrrolidone (PVP, K30), and sodium dodecylbenzenesulfonate (SDBS, 90%) were purchased from Aladdin Chemical Reagent Co., Ltd (Shanghai, China). Graphene powders (XF001W, physical method, 99 wt%, outer diameter: 0.5–5 μm, single layer ratio: 80%, special surface area: 500–1000 m² g⁻¹, and electrical resistivity ≤0.30 Ω cm) were purchased from Nanjing XFNANO Materials Co., Ltd (Nanjing, China). MWCNT powder (TNM0, CVD method, 98 wt%, outer diameter: 4–6 nm, length: 10–20 μm, special surface area: 500–700 m² g⁻¹, and electric conductivity > 100 S cm⁻¹) was purchased from Chengdu Organic Chemicals Co. Ltd (Chengdu, China). Cotton fabric (150 g m⁻² and yarn count: 40 × 40) was purchased from Suzhou Keelhuan Textile Co. Ltd (Suzhou, China).

**Synthesis of the self-cross-linkable TSiPD⁺ dispersant**

The dispersant was synthesized according to our previous report. Briefly, PDA (1.08 g, 10 mmol) was dissolved in anhydrous methanol (25 mL) and reacted with GPTMS (9.75 g, 40 mmol) under reflux for 48 h. The reaction was conducted in a dark and anhydrous environment. Subsequently, methanol was removed by rotary evaporation, and a viscous brown liquid was obtained as the product. The dispersant solution (1 wt%) was prepared by dissolving the product in a formic acid solution (0.1 mol L⁻¹) and stirring at 60 °C for 72 h.

**Fabrication of graphene- and MWCNTs-based E-textiles**

An aqueous dispersion (5 mg mL⁻¹) of graphene or MWCNTs was first prepared using TSiPD⁺ as the dispersant. Then a certain amount of nanocarbons was added into the TSiPD⁺ solution at a weight ratio of 1:0.8 (nanocarbons:TSiPD⁺), followed by tip-sonication using a JVD-650 L ultrasonic cell crushing homogenizer (Shanghai Precision Instrument Co., Ltd, China) at a power of 350 W (cycle: 3 s on and 3 s off) for 1 h. The E-textiles were fabricated via a continuous dip-coating method, as shown in Fig. 2. Cotton strips were immersed in the above dispersion for about 10 s and pulled out at a speed of.
about 12 cm min\(^{-1}\), and subsequently hung to dry at 60 °C in an oven. After all the coating cycles, the textiles were heated at 80 °C for 12 h. The cotton fabrics were rinsed with deionized water and thoroughly dried before use. The fabrication process for the graphene- and MWCNTs-based E-textiles was the same.

**Characterization**

Scanning electron microscopy (SEM) images were obtained using a Tescan Vega 3 XMU (Czech Republic) instrument. The electrical conductivity of the E-textiles was measured with an RTS-9 four-point probe meter (Guangzhou, China). Tensile tests were performed with a WDS-W universal testing machine (Chengde, China). The E-textiles were cut into standard splines with a width of 4 mm prior to testing. The washing durability test was performed by immersing a single piece of E-textile (5 cm × 7.5 cm) into an SDS aqueous solution (160 mL, 0.2 wt%) and vigorously stirring at 300 rpm. The washing time was 2 h for the first four cycles and 12 h for the fifth cycle, and finally the sample was sonicated in a water bath for 1 h. The resistance value between two points was measured with a Victor VC9808+ digital multimeter. Stretch sensing performance was determined by repeated loading/unloading on the E-textile strips, and the resistance value was monitored using a multimeter. The electric heating test was powered by disposable batteries (Panasonic, 9 V), and the temperature was monitored by a TES 1310 Type-K electronic thermometer (Taiwan, China).

**Results and discussion**

**Fabrication of nanocarbon-based E-textiles**

Continuous dip-coating is a potential approach for the large-scale production of E-textiles.\(^{19}\) Thus we chose this simple method to fabricate nanocarbon-based conductive fabrics. As illustrated in Fig. 2, a thoroughly rinsed and dried cotton strip with a width of 5 cm was continuously passed through a MWCNT dispersion, followed by drying at 60 °C in an oven. After only this single dip-coating/drying cycle, the MWCNTs-based E-textiles were obtained. The moving speed of the cotton strip was controlled at about 12 cm min\(^{-1}\). At this speed, the cotton fabric could be completely wetted by the aqueous dispersion due to the existence of numerous hydrophilic hydroxyl groups in the fibers. Nanocarbons could enter the spaces between the fibers driven by capillary forces and stay close to the fiber surface via electrostatic attraction.

Graphene and MWCNTs were positively charged after non-covalent functionalization with TSiPD\(^{+}\), and their zeta-potential values were +35.7 mV and +31.1 mV, respectively. The cotton fibers had a strong tendency to attract the positively charged nanocarbons in the first dip-coating cycle because their surfaces have a few negative charges.\(^{44}\) Fig. 3a and b show the surface SEM images of the cotton fabrics with one coating cycle. Both graphene and MWCNTs were uniformly coated on the fiber surface. The individual fibers are clearly seen, and the interspaces between them were retained without filling by additional nanocarbons. According to previous reports,\(^{45–47}\) dehydration condensation can take place between the silanol groups and hydroxyl groups of the cotton fibers, forming Si–O–C bonds. Similarly, this covalent bond was deduced to

![Fabrication of MWCNTs-based E-textile by continuous dip-coating method.](image-url)
be formed herein between TSiPD** and the cotton fiber, which caused the cotton fibers and nanocarbons to be tightly combined. Simultaneously, Si–O–Si bonds were formed within the nanocarbon layer through the dehydration condensation among the silanol groups. As a result, the nanocarbons were firmly immobilized on the fiber surfaces.

Nanocarbons can be continuously adhered to the fabrics by repeated dip-coating. Fig. 3c and d show the SEM images of the cotton fabrics after twenty coating cycles with graphene and MWCNTs, respectively. As revealed from the insets, the interspaces between the fabric fibers were filled with nanocarbons. Since the negative charges on the cotton fibers were neutralized by the positively charged nanocarbons after the first coating cycle, the hydrogen bond between the silanol groups and Si–O–Si networks should be responsible for the continuous capture of nanocarbons in the repeated cycles. Extended Si–O–Si networks were formed after each coating, which ensured the continuous increase in nanocarbons on the fabrics, and simultaneously preserved good fastness. The above approach is quite effective and easy for the fabrication of carbon-based E-textiles compared with most of the previous reports, wherein pretreatment of the substrates and pristine nanocarbons is generally employed.39,40,42,48 For example, cotton and synthetic fabrics are often boiled in alkaline media or treated with plasma to increase their surface charges42,48 and MWCNTs are usually treated by oxidation and/or further functionalization to endow their surfaces with desired groups.39,40 Besides, intermedia substances such as bovine serum albumin (BSA) are also commonly used to enhance the interaction.30,32,49 These tedious and time-consuming processes are unnecessary in our continuous dip-coating approach, demonstrating its superiority.

Fig. 4a shows the increment in fabric weight with coating cycles. The linear curves indicated that both graphene and MWCNTs were continuously adhered to the fabrics at a constant speed. After twenty coating cycles, the weight of graphene and MWCNTs per square centimeter fabric was about 1.84 mg cm⁻² and 1.88 mg cm⁻², which was specifically a 12.17 wt% and 12.46 wt% weight increment compared with the weight of pristine cotton fabric, respectively. Fig. 4b shows the photographs of the E-textiles placed on a backlight. The light transmittance was used to inspect their porosity, which also reflects their air-permeability.50,51 The backlight could penetrate the MWCNT-based textiles through the yarn gaps even after twenty coating cycles, demonstrating their good air-permeability. However, for the graphene-based textile, the transmitted light was significantly reduced after twelve coating cycles, and it became opaque after twenty coating cycles, illustrating that its air-permeability was seriously affected. The different permeabilities is most possibly caused by the different morphologies of the nanocarbons. The MWCNTs with a tube structure tended to coat around individual fibers first and then single yarns and could not exist in the large gaps between the yarns in limited coating cycles. However, for graphene, it could bridge the large gaps after several coating cycles due to its flake form. These results are consist with the SEM images in Fig. 3c and d, where the major difference lies in whether the interspaces were filled by the nanocarbons.

Electrical performance of the E-textiles

Fig. 5a shows the sheet resistances of the E-textiles with different coating cycles. The cotton fabric after one coating cycle with graphene had a sheet resistance of 128 kΩ sq⁻¹, which is sufficiently low to work as an anti-static fabric.52 However, this value was dramatically reduced by two orders of magnitude to 0.95 kΩ sq⁻¹ after the third coating cycle, and it continually decreased with more coating cycles. After twenty coating cycles, the sheet resistance was as low as 38.4 Ω sq⁻¹. Much lower values could be obtained with more coating cycles (29.8 Ω sq⁻¹ was reached after thirty coating cycles). However, the change in resistance became slow. For example, the sheet resistance only reduced from 48.3 Ω sq⁻¹ to 38.4 Ω sq⁻¹ when the coating cycle increased from fifteen to twenty despite the constant increase in graphene weight (Fig. 4a). Moreover, the air-permeability of the fabric was further reduced with more coating cycles. Hence, excessive coating cycles are not desirable.

Unlike graphene, the sheet resistance of the MWCNT-coated fabric was as low as 5.5 kΩ sq⁻¹ only after one coating cycle. This indicated that a better connected conductive network was formed by MWCNTs. However, after five coating cycles, the resistance was higher than that of the graphene-based fabric. As mentioned above, due to the large aspect ratio, the interspaces between the yarns could be gradually bridged by

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**Fig. 4**  (a) Weight increment of the E-textiles with different coating cycles and (b) photographs of the E-textiles on a backlight.
A similar morphology was observed for the MWCNT-coated fibers before (Fig. 6c) and after vigorous agitation and sonication (Fig. 6d), demonstrating they can endure harsh conditions also.

The evolution of the electrical resistance of the graphene-coated fabric prepared via two, four, or six-coating cycles with washing cycles is shown in Fig. 6e. The values were measured using a two-point method with a digital multimeter, as shown in the inset of Fig. 6f. The resistance of the sample with two coating cycles slightly increased from 56.10 kΩ to 58.30 kΩ after the washing test, while that with four coating cycles was slightly reduced from 3.28 kΩ to 3.18 kΩ. A relatively significant increment in resistance, namely, from 0.63 kΩ to 0.93 kΩ (about 47% increase), was observed for the sample with six coating cycles after the washing test. This can be interpreted as follows: as the coating cycle increased, the interspaces between the cotton fibers were gradually bridged. Thus, the fabric showed reduced flexibility and more likely cracked under friction or buckling. As a result, the electron transfer pathway was destroyed, causing an increase in resistance.

For the MWCNT-coated fabrics, there is no significant resistance change for all three samples (Fig. 6f). After the whole washing test, the resistance of the fabrics with four and six coating cycles only increased by 6.2% and 7.8%, respectively. Whereas, for the fabric with two coating cycles, the resistance value slightly decreased by 8.6% after the last sonication step. The reason for this is not clear. Maybe some of the TSiPD* molecules were not fully cross-linked and redissolved in the water during sonication, and thus reduced the blockage between MWCNTs. Actually, according to previous reports, most of the E-textiles resistance values significantly increased after the washing test. Due to the lack of strong covalent bonds to firmly immobilize the nanocarbons, the unfixed graphene or MWCNTs would be removed easily, seriously deteriorating the electrical conductivity. For example, the sheet resistance of the E-textile fabricated by the filtration of GO on the fabric and subsequent reduction increased from 0.9 kΩ sq⁻¹ to 2.0 kΩ sq⁻¹ after the washing test, which was about a 122% increase. Even worse, its sheet resistance steadily increased during the washing test.

The photograph of the textile [with four coating cycles] after the washing test is shown in Fig. 7a (top), which retained its original appearance. In contrast, the MWCNTs-based textile fabricated using PVP as a dispersant was partially bleached after the same washing test (Fig. 7a, bottom), in which the resistance also remarkably increased. To further demonstrate the good durability of our E-textiles, several pieces of fabric coated with MWCNTs for four coating cycles were immersed and subsequently sonicated in various solvents, including water, ethanol, tetrahydrofuran (THF), dichloromethane (CH₂Cl₂), and dimethylformamide (DMF). The E-textiles fabricated from PVP- and SDBS-stabilized MWCNTs were employed as the control samples for comparison. Photographs of the textiles after bath sonication for 5 min are shown in Fig. 7b. All the solvents remained clear when TSiPD* was used as the dispersant, and

**Fig. 5** (a) Sheet resistance of the E-textiles with different coating cycles and (b) MWCNT-based E-textile connected with an LED light in a series circuit, inset: textile resistance measured with a digital multimeter.

**Durability of the E-textiles**

Washability is a significant indicator to evaluate the durability of E-textiles. The nanocarbon-based fabrics were tested by vigorous agitation in 0.2 wt% SDS aqueous solution, as shown in the inset of Fig. 6f. The washing time was increased to 12 h in the fifth cycle. Finally, the fabrics were bath sonicated for 1 h, which was regarded as the sixth cycle. Fig. 6a–d show the SEM images of the graphene- and MWCNT-coated fabrics (prepared via four coating cycles) before and after the washing test. The graphene nanosheets were uniformly stacked on the fiber surface (Fig. 6a) and there was no distinct morphology change after the test (Fig. 6b), demonstrating their good coating fastness. A similar morphology was observed for the MWCNT-coated graphene and additionally contribute to the electron transfer pathway, thus resulting in a lower sheet resistance. Nevertheless, as the coating cycles further increased, the sheet resistance also continually decreased in the case with MWCNTs. The fabric after twenty coating cycles had a sheet resistance of 71.6 Ω sq⁻¹. In addition, it still possessed good air-permeability (Fig. 4b) because MWCNTs did not fill the yarns interspaces yet (Fig. 3d). A fabric (7 cm × 5 cm) with a sheet resistance of 33.2 Ω sq⁻¹ was obtained after thirty coating cycles. The resistance value of the same fabric was measured as 137.6 Ω by a digital multimeter, as shown in Fig. 5b, which was used as a connected part in a series circuit, and a small LED light was easily lit under 3 V voltage.

Graphene or MWCNTs would be removed easily, seriously deteriorating the electrical conductivity. For example, the sheet resistance of the E-textile fabricated by the filtration of GO on the fabric and subsequent reduction increased from 0.9 kΩ sq⁻¹ to 2.0 kΩ sq⁻¹ after the washing test, which was about a 122% increase. Even worse, its sheet resistance steadily increased during the washing test.

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no MWCNTs peeled off under sonication. This indicated that our E-textiles are durable in various solvents. The generated C–O–Si and Si–O–Si covalent bonds across the entire coating layer are the fundamental reason for their excellent durability. These covalent networks firmly immobilized MWCNTs on the fabric, which withstood the washing and sonication test. Actually, the solvents still remained clear even after sonication for 1 h. In contrast, the E-textiles based on PVP- or SDBS-stabilized MWCNTs were quickly destroyed in certain solvents only after 5 min. The solvents became turbid or completely opaque due to the peeling off of MWCNTs from the fabrics. The main reason for this is that the MWCNTs could not be covalently linked by the dispersant. These results strongly evidenced the excellent durability of our E-textiles.

Mechanical strength is a critical parameter to evaluate the durability and service life of E-textiles. However, for the graphene-based E-textiles fabricated via the GO route, their mechanical strength may be reduced after the reduction of GO because cotton fibers can be damaged during specific chemical processes and heating at high temperature.30,35 This issue does not exist in our approach. Fig. 8a shows the typical loading-displacement curves of the coated and uncoated cotton fabrics. All the samples were cut into standard splines along the same direction without distinguishing the warp and weft directions for testing. The loading capacity of the cotton fabrics increased by 19.0% and 22.0% after four coating cycles with graphene and MWCNTs, respectively. Moreover, the elongation at break increased by about 13.8% and 3.6%, respectively. This indicated that the fabrics retained their flexibility.

Fig. 7 Photographs of the MWCNT-based textiles (a) prepared using TSiPD⁺⁺ and PVP as dispersants after the washing test and (b) prepared using TSiPD⁺⁺, PVP, and SDBS as dispersants after sonication in water, ethanol, THF, dichloromethane, and DMF for 5 min.

Fig. 6 SEM images of the fabrics coated with graphene (a) before and (b) after the washing test and coated with MWCNTs (c) before and (d) after the washing test. Variation in the electrical resistance of (e) graphene- and (f) MWCNT-based E-textiles with washing cycles. Insets: (left) Washing process, and (right) resistance measurement with a digital multimeter.

**Stretch sensing and electric heating performance of the E-textile**

E-textiles can potentially be used as strain sensors. Thus, we studied the stretch sensing performance of our E-textiles using...
a self-made apparatus, as shown in Fig. 9. The sample was cut into 2 cm x 7.5 cm and clamped leaving it with 5 cm in length to suffer stretching. The sensing performance was indicated by the resistance change with the loading weight of 100 g, 200 g, and 500 g.

Fig. 10 shows the plots of relative resistance change (ΔR/R₀, R₀ – initial resistance of the sample and ΔR – the resistance change of the sample in the stressed or released state) as a function of loading time (s) for the E-textiles with two coating cycles. When the loaded weight was 100 g, both the graphene- and MWCNT-based E-textiles exhibited a good sensing performance. After the load increased to 200 g, the graphene-based E-textile still retained its good sensing performance; whereas, the resistance change range of the MWCNT-based textile became narrow, and its stretching sensing ability was nearly lost after fifty loading/unloading cycles. This indicated that the E-textile based on graphene has a better sensing performance than that based on MWCNTs. With a further increase in load to 500 g, the graphene-based E-textile failed after 20 loading/unloading cycles; and the resistance value of the MWCNT-based textile reversed at the stress and release state. Thus, the E-textiles with two coating cycles could not satisfy the stretching sensing performance at high load because over stretching would lead to breakage of their conductive coating, particularly the part bridging the interspaces of the fibers.

The stretch sensing performance of the E-textiles could be improved by multiple coating (Fig. 11). The graphene-based textile with twenty coating cycles showed a distinct resistance change even at 500 g load (Fig. 11c), and its sensing precision was better at all loaded weights in comparison with the sample with two coating cycles. This is because the thicker graphene layer increased the loading capacity of the E-textile, in accordance with its mechanical strength. Similarly, the MWCNT-based E-textile with twenty coating cycles also showed a much better performance at 100 g and 200 g load. However, the resistance of the stressed and released state simultaneously increased after each stretching at 500 g load (Fig. 11f), implying microstructure changes within the E-textile. It seemed that the stretching sensing performance of the MWCNT-based E-textile is inferior to the graphene-based E-textile.

Highly conductive E-textiles can also be used as a flexible electric heater. Thus, the heating performance of the fabrics coated with graphene and MWCNTs was studied, as shown in the inset of Fig. 12a. Two copper slides were used as electrodes in the opposite sides to reduce the influence of contact resistance. The heating performance was evaluated by monitoring the central region temperature of the fabric with an electronic thermometer.

Fig. 12 shows the temperature plots against time under different voltages for the MWCNT-based E-textile with a sheet resistance of 33.2 Ω sq⁻¹ and the graphene-based E-textile with a sheet resistance of 29.8 Ω sq⁻¹. When powered by a 9 V battery, the MWCNT-coated fabric quickly reached 35 °C within 90 s, which remained nearly unchanged with time. Similarly, the graphene-based E-textile also quickly reached a temperature of 40 °C within 120 s, and slowly increased afterwards. However, their heating curves were clearly different when they were powered by 18 V and 27 V batteries. The temperature for both the graphene- and MWCNT-based E-textiles first reached a steady stage within a short time, and then gradually increased to another steady stage. This phenomenon can be ascribed to
the character of the high power discharging of the disposable batteries, which can cause their internal resistance to increase and efficiency degradation. The temperature reached at 840 s was 35.2 °C, 45.3 °C, and 53.2 °C for the MWCNT-coated samples powered by 9 V, 18 V, and 27 V batteries, respectively. Under the same voltage and time, the temperature reached 43.1 °C, 57.7 °C, and 66.2 °C for the graphene-coated fabric, respectively, indicating its better heating performance resulting from its relatively lower resistance. Thus, these highly conductive E-textiles have potential applications as portable electric heaters, which can be helpful in winter to protect against the cold.
Conclusions

In summary, based on a self-cross-linkable dispersant for graphene and MWCNTs dispersions, highly conductive and ultra-durable conductive fabrics were obtained via a continuous dip-coating/drying process. Due to the dehydration condensation of the silanol groups of the dispersant, a covalent Si–O–Si network was formed across the surface conductive layer, which firmly immobilized graphene and MWCNTs on the fabric surface. Conductive fabrics with a sheet resistance as low as 33.2 Ω sq⁻¹ and 29.8 Ω sq⁻¹ were obtained by coating with MWCNTs and graphene, respectively. The fabrics possess a low sheet resistance without compromising their breathability and comfortability. After washing and sonication tests in water and various organic solvents, the resistance of the fabrics remained nearly unchanged, demonstrating their excellent durability. The E-textiles exhibited good stress sensing and electrical heating performances, suggesting their potential application as stretching sensors and portable heaters. Our study provides a basic solution for the large-scale production of highly conductive E-textiles with long service life.

Conflicts of interest

There are no conflicts to declare.

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