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Developing Graphene-based Conductive Textiles Using Different Coating Methods

Babak Abdi, Ali Tarhini, Hossein Baniasadi, and Ali R. Tehrani-Bagha*

In this research, a series of graphene-based conductive textiles is developed by three different coating methods, including dip-coating (D), airbrushing (A), and filtration (F). The cellulose substrate consists of a blend of cotton and rayon fabric, and the coating formulation is based on a mixture of graphene powder as a conductive filler, polyurethane (PU) as a binder, and poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP) as a thermoplastic co-binder. The thermogravimetric analysis (TGA) results are used to prove an enhancement in the thermal stability of the coated fabrics. The graphene content of the coated samples is also estimated from the char residue at 800 °C of the TGA profiles. The graphene-based coating converts the water-adsorbing cellulose fabric to a hydrophobic surface as the water contact angle raises from 0° to more than 107° after coating. The mechanical properties of the plain cellulose fabric enhance considerably in terms of tensile strength and tensile modulus, where the highest improvement is seen in the Dip-coating method, with an 89% increase in tensile strength compared to cellulose fabric. The graphene-based coating developed in this work enhances the physical, thermal, mechanical, and conductivity properties of the plain cellulose substrate. The resulting coated fabrics can be potentially used in wearable smart electronic textiles.

1. Introduction

Functional textiles are created with specific features and properties that go beyond their traditional uses. In addition to providing basic functions such as flexibility, breathability, and lightweight in conventional textile and clothing applications, functional textiles are designed to meet specific end-use requirements by adding some functional chemicals. These functionalities include electrical and thermal conductivity, water repellency, fire retardancy, antibacterial properties, self-cleaning capabilities, and UV protection.^[1–3]

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Graphene is a 2D honeycomb carbon lattice that has outstanding optical properties,^[4] robust mechanical stability,^[5] relatively high electrical $(\approx 10^6 \text{ S m}^{-1})$,^[6,7] and thermal conductivity (2000-6000 W m⁻¹ K⁻¹),^[6,8] excellent flame retardancy,[9] and antimicrobial properties.^[10] Thus, graphene has been used in different applications, including composites,^[11–13] sensors,^[14] capacitors,^[15] electronic devices,^[16] and filtration membranes.^[17,18] Researchers have applied graphenebased coating on different substrates to add functional properties and enhance their performance using various techniques, including the dip-coating method,^[19-21] pad-dry-cure process,^[22-24] solution casting,^[25–27] screen printing,^[28] spraying,^[4,17,18] chemical vapor deposition, and spin coating method.^[29]

Graphene has shown promising results in producing electrically conductive textiles via different coating methods. Nooralian et al. utilized a layerby-layer assembly technique to coat a

fabric, employing two distinct sprays, one containing functionalized graphene powder solution in deionized water as a functional material and the other containing a mixture of vinyl phosphonic acid as a polymerizable monomer and azo-bis-isobutyronitrile in deionized water. They applied ten coating cycles using 20 g L⁻¹ graphene dispersion and reported an electrical conductivity of 0.0245 S cm^{-1.[30]} Furthermore, Gao et al. used a dip-coating process for obtaining a conductive cotton fabric with a mixture of graphene oxide (GO) aqueous dispersion and fresh hexane solution containing dihydroxyl-terminated polydimethylsiloxane (PDMS), and tetrabutyl titanate [Ti(OBu)₄]. The GO was reduced, and a hybrid of PDMS/TiO₂ was cross-linked to the surface of the fabric in a reaction chamber containing the vapor of the mixture of hydroiodic acid (HI) and acetic acid (HAc). The reported surface electrical resistance for this sample was 76 $\Omega~\text{cm}^{-1}.^{[31]}$ Cao et al. dip-coated a silk fabric in a graphene oxide aqueous dispersion followed by the reduction of the coated fabric using sodium hydrosulfite. The surface electrical resistance of the coated silk fabric after nine cycles of repeated dipping and reducing processes was $3.2 \text{ k}\Omega \text{ cm}^{-1}$.^[32] Moreover, Ji et al. coated a silk fabric in a graphene oxide hydrosol and then reduced it in the I-ascorbic acid solution. The mean sheet resistance of the coated sample with ≈ 19.5 wt% of reduced GO was 0.13 k Ω sq^{-1.[33]} Goda et al. ADVANCED SCIENCE NEWS _____ www.advancedsciencenews.com

Table 1. Physical properties of the heat-pressed coated samples.

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Coating method	Sample code	Weight of CF [g]	Coating weight [g]	Thickness of CF [µm]	Thickness after coating [µm]
CF coated with Filtration volume of ink = 60 mL	CF-F60	0.93	1.09	317 ± 4	295 ± 12
CF coated with Filtration volume of ink = 30 mL	CF-F30	0.90	0.56	317 ± 4	222 ± 10
CF coated with Filtration volume of ink = 10 mL	CF-F10	0.92	0.18	317 ± 4	199 ± 10
CF coated with Dip-coating	CF-D	0.95	0.42	317 ± 4	190 ± 19
CF coated with Airbrushing	CF-A	0.95	0.18	317 ± 4	195 ± 12

*CF, the cellulose fabric substrate.

decorated the graphene sheets with o-carboxymethyl chitosan nanoparticles in deionized water and used the modified graphene sheets as a bio-binder for coating silver and polypyrrole on the surface of the cotton fabric via dip-coating method. The most conductive coated sample reported in their study had a sheet resistance of 0.05 k Ω .^[34] Despite using graphene as a very conductive filler in these research works,^[30-34] the resulting coated textiles did not have very high conductivity. This can be due to a) weak intermolecular interactions between graphene particles and textile substrates, b) the use of graphene oxide (GO) with more defects and lower conductivity than pure graphene, c) insulated gap between graphene particles in the coating, and d) low orientation and alignment of graphene particles in the coating. Hence, the production of highly conductive textiles remains a significant challenge, emphasizing the need for ongoing exploration of straightforward, eco-friendly, affordable, and scalable coating methods.

In this work, three different coating processes (i.e., filtration, dip-coating, and airbrushing) have been employed to coat graphene platelets on cellulose fabric to produce highly electrically conductive textiles. The rationale for selecting these specific coating methods is rooted in their distinct attributes and suitability for different applications. Dip-coating was chosen for its capacity to immerse the fabric in a coating solution, ensuring a uniform and substantial coverage that is particularly well-suited to the textile industry. Airbrushing, commonly associated with artistic applications, was favored for its ability to create a thin, precise coating layer by spraying coating ink onto the substrate. However, it's acknowledged that this method may have challenges in ensuring the uniformity of the coating on the substrate. In contrast, filtration, a relatively unconventional method, was employed for its efficiency and resource optimization. It relies on a filter to separate the coating material from a solvent, followed by its deposition onto the fabric.

In this respect, different physical-chemical properties of the fabricated conductive textiles were investigated and discussed to prove the benefit of the employed methods in developing flexible conductive textiles with enhanced properties compared to the plain cellulosic substrate. This study sheds more light on the topic and provides valuable information for fabricating conductive textiles, which can open a window to develop conductive fabrics with significant potential in wearable smart electronic textiles.

2. Results and Discussion

2.1. Fabrication and Structural Characterization

After several screening trials, a coating mixture of graphene, a polyurethane (PU)-based binder, and PVDF-HFP was utilized. PVDF-HFP, as a thermoplastic hydrophobic copolymer, was employed to enhance the binding of the coating formulation. Furthermore, it was realized that the heat-pressing enhanced the uniformity and conductivity of the coated samples. **Table 1** displays the physical properties of the coated textiles. The samples coated using the filtration method exhibited a higher weight gain, which was directly proportional to the filtration volume. Additionally, there was a clear positive correlation between the coating weight and the thickness of the coated samples, indicating that higher coating weights resulted in thicker coatings.

The chemical structure of the pure fabric, as well as the coated samples, was investigated by ATR-FTIR spectroscopy analysis. The spectra are illustrated in Figure 1. The spectrum of the CF presents peaks between 3500-3000 cm⁻¹ corresponding to the hydroxy groups of cellulose, two peaks at 2962 and 2896 cm⁻¹ attributing to the asymmetrical and symmetrical stretching of CH₂, two bands at 1652 and 1264 cm⁻¹ originating to C=O stretching, a peak at 1430 cm⁻¹ assigning to symmetric bending of CH₂, a peak at 1158 cm⁻¹ arising of the anti-symmetrical bridge of C–O–C, two bands at 1057 and 1020 cm⁻¹ attributing to stretching of C–O, a peak at 892 cm⁻¹ originating to β -linkage of cellulose, and a peak at 670 cm⁻¹ assigning to out of plane bending of OH.^[35] The spectra of coated samples revealed the rise of new peaks, indicating a different surface chemistry compared to neat CG. For instance, a peak appeared at 1725 cm^{-1,} which was assigned to the typical C=O absorption band of the PU linkage,^[36] suggesting the existence of the polyurethane-based

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Wavelength (cm-1)

Figure 1. a) FTIR spectroscopy analysis of the neat and coated fabrics prepared using different coating methods, b1) CF, b2) CF-F60, b3) CF-D, and b4) CF-A samples characterization peaks.



Figure 2. SEM images of a) CF, b) CF-F60, c) CF-D, and d) CF-A.

binder. Furthermore, a new band was formed at 765 cm⁻¹, which was assigned to α phase structure of PVDF-HFP.^[37] These peaks confirm the successful deposition of the coating materials on textile substrates. As expected, the ATR-FTIR spectra of the coated samples showed similar graphs for CF-F60, CF-D, and CF-A samples.

Figure 2 shows the front view SEM micrographs of the untreated and coated fabrics. The twisted fibers of the untreated knitted fabric are visible, illustrating a clean surface. The surface of the coated fabrics, on the other hand, shows the distribution of the graphene flakes. However, significant differences can be detected in the fabrics coated with different coating methods. For instance, the surface of the CF-F60 sample presents a large number of graphene flakes stacked on top of each other and well connected with a binder. However, the surface of the CF-D fabric shows a lower amount of graphene flakes compared to the CF-F60 one. Furthermore, the SEM micrograph shows a lower amount of graphene flakes with poor interconnections on the surface of the CF-A sample compared to the surface of the CF-F60 and CF-D samples. It seems in the filtration method, due to the slow rate of the filtration process, the graphene flakes stack on each other very slowly with the help of gravitational force, allowing graphene flakes to be aligned sitting on top of each other, which helps to form a well-connected and in-plane accumulation of the graphene flakes. In contrast, in the dip-coating process, the uptake of the coating mixture is limited on the fabric. Likewise, in the airbrush spraying, due to the speed of the coating procedure, the graphene flakes spread on the fabric in a disordered pattern that causes the formation of graphene islands, which results in the poor interconnection of graphene flakes in the CF-A sample. The cross-section SEM images of the CF-F60, CF-D,

and CF-A samples were used to further investigate the quality of the coating layer. The micrographs are depicted in **Figure 3**. It can be seen that for the CF-F60 sample, the graphene flakes were stacked on top of each other uniformly, and they were wellconnected with the help of binder and PVDF-HFP. However, the cross-sectional images of CF-D and CF-A showed a random orientation of graphene flakes in the coating layer. In addition, a lower amount of graphene flakes can be seen for the CF-A sample compared to CF-D and CF-F60. Furthermore, the coating layer thickness measured from the cross-section SEM images (Figure 3a,b) for CF-F60 and CF-D varied in the range of 100– 120 µm and 50–70 µm while for the CF-A sample, a clear line between the coating layer and fabric was not seeable.

The surface characteristic of the sample in terms of water wettability was examined by water contact angle measurements. Figure 4 shows the water contact angle deposited on the CF, CF-F10, CF-F30, CF-F60, CF-D, and CF-A surfaces after 10 s. CF has a porous hydrophilic substrate in which the water droplet gets absorbed in it immediately. In contrast, the CF-F10, CF-F30, CF-F60, CF-D, and CF-A samples show a water contact angle of higher than 100 °, indicating a hydrophobic surface.^[38] More importantly, the water contact angle was constant even after 30 s, suggesting a durable hydrophobic surface after coating. The observed hydrophobicity could be attributed to the hydrophobic nature of both GNPs and PVDF-HFP in the coating formulation.^[39,40] The WCAs for CF-F10, CF-F30, and CF-F60 consistently measured between 100 and 108 degrees, confirming the hydrophobic effectiveness of the coating ink, even when used with reduced coating volumes.

The effect of the coating layer on the thermal stability of the neat CF fabric, as well as the amount of coated layer, was



Figure 3. SEM images of the cross-section of the a) CF-F60, b) CF-D, and c) CF-A.

investigated by thermal gravimetric analysis (TGA). The TGA and DTG curves of the samples are shown in Figure 5a,c. Similar to the literature,^[41-46] the neat cellulose fabric presented a single decomposition trend upon heating under a nitrogen atmosphere. This decomposition occurred in the range of 300-400 °C, corresponding to decarboxylation and decomposition of crystalline parts of cellulose.^[30,47] The sharp peak in the DTG curve at 345 °C was due to this decomposition stage and presented the maximum degradation rate temperature (T_{max}). It should be noted that a minor initial weight loss of nearly 5% could also be seen between 70 and 120°C, attributed to the removal of absorbed water. A minor peak in the DTG curve appeared due to this decomposition step. After the complete thermal decomposition, the char was created at 800 °C as the final remanent. The TGA and DTG curves of the coated samples revealed a significantly different decomposition behavior overheating. Namely, two decomposition regions accompanied by two DTG peaks could be seen in the thermograms, suggesting the degradation of the neat cellulose fabric as well as the binder and PVDF-HFP, and it is visible in the DTG curves of the samples with two new $T_{\rm max}$ values of 385–395 °C and 460–470 °C. Figure 5b,d shows the TGA and DTG curves of the PVDF-HFP and the binder, where $T_{\rm max}$ of 398 and 443 °C were seen for the binder and PVDF-HFP. The char residues for CF-F60, CF-D, CF-A, and CF samples were determined to be 40.6%, 22.2%, 16.35%, and 9.58%, respectively. The char residue observed in the samples can be attributed to their graphene content.

Since graphene particles are responsible for making textiles conductive, it would be important to estimate the graphene loading in the formulation. The char residue was used to determine the approximate graphene content of different coated samples. This was performed by comparing the char residue of these www.advancedsciencenews.com

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Figure 4. The water contact angle of a) CF, b) CF-A, c)CF-D, d) CF-F10, e) CF-F30, and f) CF-F60.



Figure 5. TGA and DTG curves of CF, CF-F60, CF-F30, CF-F10, CF-D, CF-FWG, CF-A, PVDF-HFP, and the binder. CF-FWG is the sample coated with the binder without graphene.

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Sample

CF-FWG

CF-F60

CF-F30

CF-F10

CF-D

CF-A

CF

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Ultimate strain^{*} [%]

49.30 ± 4.71

45.22 ± 1.53

34.11 ± 7.37

 36.76 ± 0.88

29.33 + 5.73

32.08 ± 2.26

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 Table 2. Char residue and approximate graphene content in coated samples.

Char residue [%]

9.58

114

39 97

24.45

19.22

22.21

16 36

	fabrics.		
Approximate graphene [%]	Sample	Tensile modulus [MPa]	Tensile strength* [MPa]

4.93 ± 0.46

70.27 + 22.29

360.70 ± 30.87

50.18 ± 15.12

359.73 + 43.28

287.20 ± 36.89

*These values were extracted after the force reached 18 N

CF

CF-A

CF-D

CF-F10

CF-F30

CF-F60

0

0

28 57

13.05

7.82

10.81

4 96

samples with the char residue of CF-FWG and CF. Table 2
presents the char residue and graphene content data for CF-
F60, CF-F30, CF-F10, CF-D, and CF-A. The difference in char
residue between CF-FWG and CF was 1.82%. Considering the
char residue percentage of PVDF-HFP and PU-based binder
(Figure 5b), one can conclude that the amounts of PVDF-HFP
and PU-based binder in the coating formulation were ≈1.45%
and 0.37%, respectively. It should be highlighted that it was as-
sumed that the char residue percentage of PVDF-HFP and PU-
based binder was the same for all coated samples due to the
identical coating formulation used for all samples. Therefore, the
difference in char residue between the coated samples and CF-
FWG provides an approximate measure of the graphene content
in each sample.

2.2. Mechanical Properties

Flexibility is a great concern in designing wearable smart electronic textiles because the conductive fibers need to maintain stable functionality under complex and harsh mechanical deformations of the human body.^[48] Thus, tensile testing was performed to investigate the effect of coating on the stretchability and mechanical stability of the developed conductive textiles. All the samples were stretched up to 18 N with a rate of 0.5 N min⁻¹. The typical stress-strain curves are plotted in **Figure 6a**. Furthermore, the relevant mechanical properties, including tensile modulus, tensile strength, and ultimate strain, were summarized in **Table 3** and compared in Figure 6b. The neat CF fabric presented the highest strain, while it possessed the lowest tensile modu-



Table 3. Mechanical properties of the plain cellulose fabric and the coated

 9.43 ± 0.53

 17.17 ± 0.80

17.68 ± 2.02

15.72 ± 1.18

15.82 + 0.37

 11.50 ± 0.54

The different coating methods provided different mechanical properties. The breaking force of coated textiles remained practically unchanged in the airbrushed and dip-coated fabrics, whereas it tended to decrease slightly in the filtrated samples, which have already been proven to have a thicker coating. Similar results were reported for coated woven cotton fabrics in which a reduction was observed in the mechanical properties of the coated fabrics as the coating amount increased.^[50] It seems that in the filtration method, most of the coating mixture remained on the surface of the fabrics, while in the dip-coating and airbrushing techniques, the coating could, to some extent, penetrate the fibrous substrate and, as a result, an additional layer was formed on the fiber surface by a coating of polymers and graphene particles.^[51] It should be highlighted that within the



Figure 6. a) The typical stress-strain curves and b) the comparison between different mechanical properties of the neat cellulose fabric and the coated fabrics.

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Table 4. Electrical properties of coated fabrics.

Sample	Mean sheet	Electrical		
	resistance [Ohm sq ^{-1]}	conductivity [S m ⁻¹]		
CF-F60	2.7 ± 0.3	1337 ± 234		
CF-F30	11.3 ± 3.9	445 ± 136		
CF-F10	381 ± 133	14.8 ± 4.7		
CF-D	504 ± 133	11.1 ± 2.6		
CF-A	10884 ± 4729	0.6 ± 0.2		

tensile testing, the coating layer of the CF-F fabrics broke while the coating was preserved well in CF-A and CF-D conductive textiles, indicating the force was mainly distributed in the coating rather than the fibers due to relatively thicker coating achieved in this coating method.

2.3. Electrical Conductivity Measurements

Electrical conductivity is one of the main features in designing smart textiles for wearable electronics applications.^[52] Hence, the electrical conductivity of the neat cellulose fabric, as well as the coated samples, was examined. The results are shown in **Table 4**. The measured value for the neat cellulose fabric was 2.78×10^{-9} S m⁻¹, indicating an intrinsic electrical insulator.^[53] However, the coated samples exhibited a substantial increase in conductivity, confirming that the coating remarkably enhanced the electrical properties of the substrate fabric.

To assess the conductivity of the coated samples, they were connected to a power source along with a small diode for visualization. The test results indicated that the CF-F60 sample exhibited higher conductivity compared to the CF-D sample. This was evident as the diode lit up brightly even at very low voltages (**Figure 7**a), indicating a strong flow of current. The higher electrical conductivity in CF-F60 was attributed to the uniform and compact graphene coating formed on the surface of cellulose fabric through the filtration method, as confirmed by SEM

www.advmattechnol.de 1800 Mean Electrical Conductivity 1500 62,293x - 427,65 $R^2 = 0.9936$ 1200 (S/m) 900 600 300 0 10 20 30 0 Graphene content (%)

Figure 8. Graphene content effect on mean electrical conductivity.

images. It should be highlighted that this sample, which had the thickest coating layer, provided excellent flexibility and was restored to its original state after folding and crimping, as depicted in Figure 7b. To assess the durability of our coated samples, we subjected a CF-F60 sample to 100 bending cycles, as demonstrated in Video S1 (Supporting Information). The electrical conductivity measurements taken after 50 and 100 repeated bending cycles (see Figure 7c) showed a slight reduction of 7% and 10% in the sample's average electrical conductivity, respectively. This decrease may be attributed to the possible breakage of conductive pathways within the coating layer. Nevertheless, even after 100 cycles, the sample maintained a high level of conductivity, showing the flexibility and resilience of our metal-free conductive textiles, particularly suited for eTextile applications.

Based on the results of electrical conductivity measurements presented in Table 4, the samples were ranked in the order of CF-F > CF-D > CF-A. Figure 8 provides a clear correlation between the mean electrical conductivity results of CF-F60, CF-F30, and CF-F10 and their respective graphene content. The



Figure 7. a) Photograph of CF-F sample with high electrical conductivity in an electronic circuit, b) Surface appearance and flexibility of CF-F sample, c) Mean electrical conductivity change after bending cycles of CF-F60 sample (Video S1, Supporting Information).

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Table 5. Mean sl	neet resistance	comparison of th	is work with	previous studies	s on g	raphene-based	coating

Filler	Substrate	Methods of synthesis	Mean sheet resistance	Refs.
Graphene flakes	Cotton/Rayon	Filtration	$2.7 \Omega sq^{-1}$	This work
Graphene nanosheets	Cotton	Dip and dry	$7 \Omega \text{ sq}^{-1}$	[57]
Graphene flakes	Poly-cotton	Pad-dry-cure method	11.9 Ω sq ⁻¹	[58]
Graphene nanosheets	Lyocell	Dip and dry	40 Ω sq ⁻¹	[59]
Reduced graphene oxide	Silk	Dry-coating	$130 \ \Omega \ sq^{-1}$	[33]

CF-F60 sample, which had the highest graphene content, exhibits the highest mean electrical conductivity values. Although CF-D had a higher graphene content than CF-F10 (see Table 2), its electrical conductivity values were considerably lower. This indicates that the filtration process employed in the preparation of CF-F samples resulted in more effectively formed graphene conductive channels than the dip-coating method used in the preparation of CF-D samples. The filtration process promoted superior in-plane stagnation and alignment of graphene flakes, which exerted forces that enabled the flakes to be more effectively stacked on top of each other. This stacking resulted in the formation of continuous channels that facilitate the transfer of electrons across the coating layer, as well as a decrease in the contact resistance among the layers across the coating layer. Both of these factors play an essential role in improving the electrical conductivity of the samples.^[37,54] Moreover, the better orientation of graphene flakes due to the filtration process resulted in a higher aspect ratio for graphene nanoflakes (GNFs) in the coating layer, which significantly affected the electrical conductivity of the samples.[37,55,56]

The mean sheet resistance results of this work were compared to previously reported findings (see **Table 5**). To the best of our knowledge, the mean sheet resistance of the CF-F60 sample ($2.7 \Omega \text{ sq}^{-1}$) was considerably lower than the lowest reported mean sheet resistance ($7 \Omega \text{ sq}^{-1}$) in previous studies.^[57] In addition, this work presents a straightforward method for coating a high content of graphene on fabric compared to previous methods, which consisted of numerous dipping and drying methods.

3. Conclusions

In this study, we optimized the coating formulation and employed three different coating methods to fabricate conductive cellulosic fabric. We conducted a comprehensive evaluation of the structure and functionality of the coated samples. Our findings from FTIR analysis revealed that the coating formulation remained consistent across all samples. However, FE-SEM analysis showed that the filtration process resulted in superior in-plane alignment of graphene nanoplates. The coating positively impacted the tensile strength and tensile modulus of the cellulose fabric without noticeable changes in the tensile strain. The results of the water contact angle measurement indicated a transformation from the water-absorbing fabric to a hydrophobic surface after the application of the graphene coating.

The coated fabric with the filtration process had the highest thermal stability and residual mass compared to the other coated samples. The electrical conductivity of the filtration-based coated sample surpassed that of the dip-coated and airbrushed samples, even at lower filtration solution concentrations. The filtration process is a promising method for producing flexible, highly conductive, and thermally stable graphene-coated textiles. These findings have significant potential for the development of wearable smart electronic textiles.

4. Experimental Section

Materials: Graphene flakes with a specific surface area $\leq 40 \text{ m}^2 \text{ g}^{-1}$, purity of 98.5%, and lateral particle size $\leq 7 \mu \text{m}$ were purchased from Graphene Laboratories Inc. Polyvinylidene fluoride-co-hexafluoropropylene (PVDF-HFP) with an average molecular weight of 455 000 g mol⁻¹ and *N*,*N*-dimethylformamide (DMF) with 99.8% purity were purchased from Sigma–Aldrich (Germany). A polyurethane (PU) based binder (A-5001, Aroma-free, and hydrocarbon-rich weighted paste) was obtained from Wennström Company (Finland). A knitted fabric (175 g m⁻²) made of 70% percent rayon and 30% organic cotton with a yarn count of 60 Tex was received from the Coveross Company (Finland).

Fabrication of Graphene-Based Textile—Coating Ink Formulation: 20 g L^{-1} of graphene dispersion was prepared by mixing 0.5 g of graphene flakes in 25 mL of DMF by sonication at 35 °C for 30 min. Twenty-five milliliters of PVDF-HFP in DMF solution (20 g L^{-1}) was prepared separately by dissolving the polymer in hot DMF under constant stirring. Then, the graphene dispersion was added slowly to the PVDF-HFP solution and mixed for 15 minutes. After that, 10 mL of binder paste (A-5001) was added slowly to the DMF solution containing the GNP and PVDF-HFP, and the final solution was mixed for 15 min. Finally, the resulting ink was used to coat the cellulose fabric via three different coating methods, e.g., filtration, dip-coating, and airbrush spraying.

Fabrication of Graphene-Based Textile—Coating Methods:

- (a) Filtration Process: Filtration was performed using a Buchner funnel with a diameter of 8 cm. First, a filter paper with a particle retention of 2 μ m was placed at the bottom of the funnel, followed by a layer of the fabric to be coated. The coating ink dispersion was poured slowly onto the fabric surface and allowed to be filtered through the layers, ensuring an even distribution of the coating material.
- (b) Dip-Coating Process: The fabric was immersed in the coating suspension for 3 min, ensuring the entire surface area was fully covered. Subsequently, the fabric was placed inside an oven and dried at a temperature of 80 °C for 5 min to facilitate the adhesion of the coating onto the textile. This dipping and drying process was repeated five times to ensure a consistent and durable coating on the fabric.
- (c) Airbrush Spraying: A professional complete airbrush system (Model: ABEST, Manufacturer: Ediyworld) equipped with a 0.2 mm diameter nozzle was employed to spray the coating dispersion onto the fabric. The sample was held vertically, and the airbrush was positioned at a distance of 5 cm from the surface. The spraying process was repeated five times for the fabric's front and back sides to ensure thorough coverage.

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Fabrication of Graphene-Based Textile—Curing and Heat Pressing After Coating: The coated fabrics were dried and cured in an oven at 150 °C for 5 min after the coating process. To ensure maximum coating adhesion onto the fabric, a hot-pressing step was performed using a FONTUNE hydraulic laboratory press (Vlaardingen, Holland) for all coated fabrics. The coated samples were sandwiched between two protection films and pressed at a temperature of 150 °C and a pressure of 150 kN for 4 min. The hot pressing was accompanied by cold pressing for 10 min at the same pressure to maintain the integrity of the coating. The pure cellulose fabric sample was designated as CF (Cellulose Fabric), while the coated samples prepared via different methods were named as follows: CF-F (Coated Fabric via Filtration method), CF-D (Coated Fabric via Dip-coating), and CF-A (Coated Fabric via Airbrushing method). Additionally, CF-F10, CF-F30, and CF-F60 represent the filtered samples prepared using 10, 30, and 60 mL of the coating solution, respectively. CF-FWG is a benchmark sample representing a filtered fabric with a coating formulation that does not contain graphene.

 $\label{eq:characterization} \begin{array}{l} \mbox{Characterization} \mbox{--Thickness:} & A thickness tester Lorentzen & Wettre SE \\ \mbox{250 D} (Finland) with a micrometer resolution was used for measuring the textile thickness with an indication error of <math display="inline">\pm 1\,\mu m$ or 0.1% reading.

Characterization—*ATR-FTIR*: The Attenuated Total Reflection Fouriertransform infrared (ATR-FTIR) spectroscopy measurement was done using FT-IR Spectrometer (PerkinElmer, USA) at room temperature with LiTaO₃ (Lithium tantalate) MIR (Mid-infrared) detector with a signal to noise ratio (SNR) of 9.300:1. The spectra were recorded from 4000 to 500 cm⁻¹, with a resolution of 4 cm⁻¹.

Characterization—Scanning Electron Microscopy (SEM): The microstructure of the fabrics was studied and investigated using emission scanning electron microscopy (SEM; Zeiss Sigma VP, German) with an InBeam detector at an accelerating voltage of 20 KV. The graphene-based textile specimens were mounted on aluminum stubs using adhesive carbon tabs. A gold/palladium (80 Au/20 Pd) deposition was applied only on the CF sample before scanning.

Characterization—Thermal Gravimetric Analysis (TGA): The thermal gravimetric analysis was investigated using the simultaneous thermal analyzer NETZSCH STA 449 F3 Jupiter (Germany). A sample weight of \approx 10 mg was put inside an Alumina (Al₂O₃) crucible, and the test was done in the range of 40–800 °C with a heating rate of 10 °C min⁻¹ under a nitrogen atmosphere.

Characterization—Electrical Conductivity: The electrical conductivity for each coated fabric was measured using a four-point probe system from Ossila (UK). The change in the voltage across the inner two probes is measured when the current is passed between the outer two probes. Electrical conductivity (σ) was calculated using Equation (1).

$$\sigma = \frac{ln2}{\pi} \frac{l}{\Delta V} \left(\frac{1}{d}\right) \tag{1}$$

where d, l, and ΔV are the textile thickness, electrical current, and measured voltage difference, respectively. The reported values represent the mean of ten measurements.

Characterization—Contact Angle Measurement: The water contact angle measurements were conducted using a Theta Flex optical tensiometer from Biolin Scientific to investigate the surface water wettability. A 5.0 μ L water droplet was placed on the sample surface with a drop rate of 2.0 μ L s⁻¹. The measurement was initiated as soon as the droplet was positioned, and the imaging was done for 60 s at a rate of 1 fps.

Characterization—Tensile Testing: Different mechanical properties of the fabrics, including tensile modulus, tensile strength, and ultimate strain (%), were examined by tensile testing. The test was performed on a DMA device model Q800 with a preload of 0.1 N at a fixed temperature of 25 °C. The load rate was set as 0.5 N min⁻¹, and the sample was stretched up to 18 N. The reported values are the average of three replicates.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

coating, conductive textile, electrical conductivity, graphene, hydrophobicity, thermal stability

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