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Synthesis of PEDOT:PSS Solution-Processed Electronic Textiles for Enhanced Joule Heating

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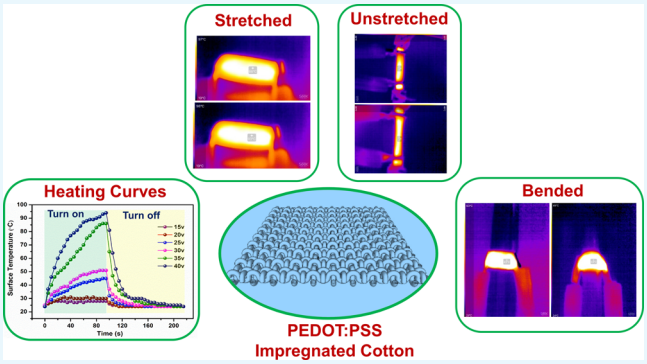
ABSTRACT: Textile-based flexible and wearable electronic devices provide an excellent solution to thermal management systems, thermal therapy, and deicing applications through the Joule heating approach. However, challenges persist in designing such cost-effective electronic devices for efficient heating performance. Herein, this study adopted a facile solution-processed strategy, "dip-coating", to develop a high-performance Joule heating device by unformly coating the intrinsically conducting polymer (CP) poly(3,4-ethylenedioxythiophene)-poly-(styrenesulfonate) (PEDOT:PSS) onto the surface of cotton textiles. The structural and morphological attributes of the cotton/CP mixture were evaluated using various characterization techniques. The electrothermal characteristics of the cotton/CP sample included rapid thermal response, uniform surface temperature distribution up to 94 °C, excellent stability, and endurance in heating performance under various mechanical deformations. The real-time illustration of the fabric heater affixed on a human finger has demonstrated its outstanding potential for thermal therapy applications. The fabricated heater may further expand it purposes toward deicing, defogging, and defrosting applications.



1. INTRODUCTION

Lightweight, high flexibility, and intelligent performance electronic devices have received immense attention in flexible electronics. Fabric-based smart materials have been considered to be emerging electronic devices that meet the requirements of flexible electronics and are compatible with multifunctional wearable electronics. These smart textile devices have been used in various platforms for energy storage,[1,2] flexible sensing,[3] thermal management,[4−9] real-time healthcare monitoring,[10−13] textile robotics,[14] protective textiles,[15,16,4,17] and intelligent clothing.[18,18−20] Typically, traditional textiles are inherently electrically nonconductive and therefore not ideally suitable for electronic applications. To date, to impart electrical conductivity, textile materials are being functionalized with enormous nanomaterials, particularly 0D metal nanoparticles,[21−25] 1D carbon nanotubes (CNT),[26] 2D graphene[27−32] and MXene,[33−36] and conducting polymers such as poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS),[37,38] and polypyrrole.[39−41] The exploration of these nanomaterials has enabled smart-textile devices for profound interests toward multifunctional electronic applications.

One of the key applications of smart-textile-based functional devices is thermal management, a technology encompassing the generation, control, and dissipation of heat from electronic devices. Such electronics devices are extensively important in managing indoor heat[42] in addition to defrosting and deicing,[43] and are also an excellent means of protection from extreme cold environment as high-altitude garments.[44] Furthermore, these devices have intriguing features that can be employed in wearable thermal therapy devices characterized as Joule heaters. Significant research has been made in this field by exploiting fabric-based electronic devices.[45] However, the efficient heating capability, safe operating voltage, and reliability of these devices hinders their continuous and long-term applications. For example, a textile-based heating element prepared by Souri et al. using a combination of a graphene nanoplatelet and carbon black showed a promising heating efficiency.[46] However, it required a large driving potential of 50 V to achieve the surface temperature of 95.3 °C. Similar heating electrodes were prepared from graphene/water-born

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polyurethane (WPU)/polyester[47] and graphene/WPU/*para*-aramid[48] composites, which required high driving voltages as prerequisites to obtain surface temperatures of ∼71 and ∼55 °C, respectively. Although such devices may be utilized for deicing or defrosting purposes, they are not suitable in terms of wearable electronic applications for thermal management or therapy. This necessitates the exploration of fabric-based electrodes for efficient and reliable heating operations.

A series of nanomaterial-enabled electrical heating devices have been constructed by integrating them with pristine textile substrates, leading to flexible smart-textile structures embedded with electronic properties. When applied as a Joule heater, they exhibited promise in their rapid thermal response, uniform heating and surface temperature distribution, excellent stability, and safe operating voltage.[49] However, owing to the risk of rigidity, the tendency of oxidation, the lack of flexibility, and the processing difficult, the utilization of metal- and carbon-based materials for smart-textile manufacturing has been restricted.[37] In contrast, PEDOT:PSS, among other conducting polymers, with its high dispersibility in different solvents, superior electrical conductivity, and excellent chemical and electrochemical stability is making strides in smart-textile development for wearable electronics.[50] Importantly, PEDOT:PSS makes textile functionalization feasible, which is ascribed to its excellent solution processability. In this context, a PEDOT:PSS-containing solution can be directly applied to the surfaces of textile materials by a simple dip-coating approach, eliminating the need for complex instrumentation during vapor-phase or in situ polymerization. The impregnation of PEDOT:PSS in textile structures results in performance augmentation by serving as either an electrical conductive unit or a gluing agent in the composite structure. For example, our previous report illustrates that PEDOT:PSS not only improves the electrical conductivity as "electrical adhesives" but also serves as a binder to prevent the leaching of graphene nanoparticles from the composite surfaces.[37] Hence, a consideration of the intrinsic characteristics of PEDOT:PSS toward the effective functionalization of smart textiles for electrical applications is worth further exploring.

This study reports a facile technique to develop a conducting polymer (PEDOT:PSS) impregnated smart-textile-based electrical heating device for wearable Joule heating. The as-obtained textiles were characterized via SEM, FTIR, and XRD to evaluate their structural and morphological characteristics. A series of electrical and electrothermal analyses were executed to understand the electrical and Joule heating performance of the composite textiles. The excellent synergies between the conducting polymer and the textile substrates resulted in high-performance electronics textiles, which showed promise in wearable Joule heating and might be useful in deicing and defrosting systems.

2. MATERIAL PREPARATION

2.1. Materials. Knitted, scoured, and bleached single-jersey 100% cotton fabrics were supplied by Texeurop (BD) Ltd. (Bangladesh). The conducting polymer (CP) PEDOT:PSS dispersion (2−3% in H_2O) was procured from Sigma-Aldrich (United States). Analytical-grade reagents, including polyethylene glycol (PEG) (M_w = 1000) and sodium hydroxide pellets (NaOH, 97% purity), were purchased from Merck, Germany. Throughout the experiments, deionized (DI) water was used.

2.2. Fabrication of PEDOT:PSS-Impregnated Textiles. Cotton textiles were first cut into pieces (specimen size of 6 × 6 cm^2) and mercerized by dipping them in a 5% aqueous NaOH solution for 15 min, then rinsed several times and dried in a flat air dryer machine (at 60 °C). The conductive coating of the polymer was deposited on the cotton textile by the simple-solution processed "dipping and drying" approach. Briefly, 1 vol % PEG-1000 was dissolved in DI water. To the PEG solution was added a previously dispersed solution of PEDOT:PSS of the desired concentration. Then, the conductive solution was magnetically stirred for 12 h, followed by probe sonication for 30 min. After that, the mercerized cotton textile was dipped for 2 min in the conducting polymer solution and dried in a vacuum chamber at 70 °C. A number of dipping and drying cycles were performed for the cotton textiles. A series of conductive textiles were fabricated via this procedure, and the specimens were marked as cotton/CP-(x) based on the number of deposited layers on the textiles, where x indicates the number of deposition cycles, i.e., x = 10 and 20. The mass loading of the dip-coated samples was measured to be ∼1.2 and 2.5 mg cm^{-2} for samples dipped for 10 and 20 cycles, respectively.

3. CHARACTERIZATIONS AND MEASUREMENTS OF MATERIALS

The X-ray diffraction patterns of the pristine cotton textiles and cotton/CP were analyzed with an X-ray diffractometer (Rigaku Ultima IV, Japan). The diffraction pattern of samples was obtained using Cu Kα radiation (λ = 0.1539 nm) with an operating voltage of 40 kV and an applied current of 40 mA. The 2θ angle ranged from 5° to 30° with a scanning rate of 4° min^{-1}. The FTIR spectra of the pristine cotton textile and cotton/CP were measured in the wavelength range of 4000−650 cm^{-1} using a Nicolet 6700 FTIR analyzer (Thermo Fisher Scientific, United Staes) in ambient conditions. An average of 64 scans with a resolution of 4 cm^{-1} were used in the attenuated total reflectance (ATR) mode. The surface and cross-sectional morphology of the untreated cotton fabric and cotton/CP were analyzed using a SEM (ZEISS Evo 18, special edition).

The electrical performance of the cotton/CP sample was obtained using a customized gold-plated four-probe resistivity meter. The sheet resistance of the composite fabrics was examined at five various positions, and the average sheet resistance was reported. The electrothermal characteristics and thermal images of the samples were determined by an infrared thermal camera (SEEK) at various applied potentials, and the average value was reported.

4. RESULTS AND DISCUSSIONS

4.1. Structural and Morphological Analysis. Various characterization techniques have been employed to reveal the structural and morphological attributes of the samples. Figure 1 shows the SEM images of pristine cotton textiles and PEDOT:PSS -reated textiles. The pristine fabrics have smooth surfaces, and no particles are apparent on the fiber surfaces (Figure 1a). A highly uniform and conformal wrapping layer of the conducting polymer was obtained on all the fiber surfaces coated with PEDOT:PSS. As shown in Figure 1b, at the lower dipping cycles of CP, a relatively less interconnected structure was formed, while higher dipping cycles of CP facilitated the formation of enhanced the interconnected networks (Figure

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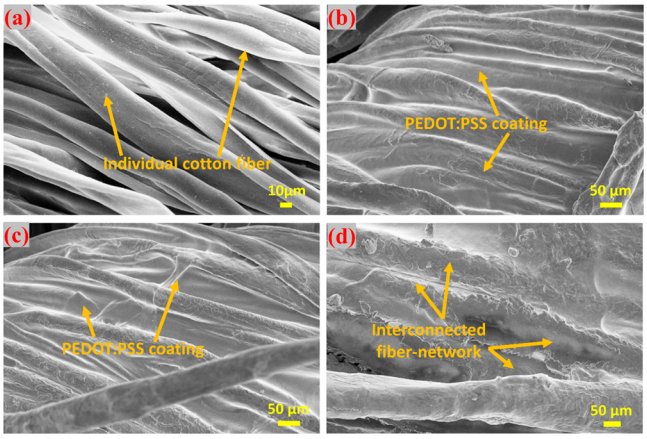

Figure 1. Morphological characteristics of pristine and functionalized textiles. SEM image demonstrating (a) scoured and bleached cotton fabric, (b) fabrics treated with lower dipping cycles of PEDOT:PSS (cotton/CP-10), and (c) fabrics treated with higher dipping cycles of PEDOT:PSS (cotton/CP-20). (d) SEM image of PEDOT:PSS-treated textiles (cotton/CP-20) at higher magnification. Arrows show the interconnected fiber networks.

1c). This consequently imparted an efficient conductive pathway, as observed from the higher-magnification of SEM image shown in Figure 1d. Overall, this could be due to the strong interfiber bonding among the adjacent fibers and PEDOT:PSS that emerged from the presence of PSS as a cross-linker.

The corresponding XRD patterns of pristine cotton and PEDOT:PSS-treated textiles are demonstrated in Figure 2a. The pristine cotton revealed four characteristic diffraction patterns at distinctive reflection planes at 2θ = 14.45° (110), 2θ = 16.33° (1$\underline{1}$0), 2θ = 19.16° (120), and 2θ = 22.68° (200). These reflections reveal that the pristine cotton textile is composed of the cellulose-I crystalline polymorph.[51] The sample treated with a low PEDOT content (cotton/CP-10) has overlapping diffraction patterns identical to those of pristine cotton, with corresponding lattice spacings (d) of 6.12, 5.43, 4.64, and 3.98 Å for planes 110, 110, 120, and 200, respectively. However, no peak was observed at 25° (plane 020) for the diffraction pattern of PEDOT:PSS, as it merged within the broad peaks of the cotton textiles (200). Increasing the content of PEDOT:PSS on the fabric's surface (cotton/CP-20) led to a remarkable increase in intensity along with enhancements in both the sharpness and the widths of X-ray peaks. Moreover, it is noted that PEDOT:PSS treatment on the cotton fabric modifies the stereoregularity of the cellulose matrix, thus increasing the polymer chain lengths and the interaction of hydrogen bonds. This suggests structural synergies between the cotton textiles and PEDOT:PSS, which might facilitate conductive pathways in the composite textile structure. This is further supported by the investigation of the effects of PEDOT:PSS treatment on textiles performed by Alamer et al.[52]

Figure 2b shows the FTIR spectra of the pristine cotton and PEDOT:PSS-impregnated cotton. Pristine cotton exhibits two distinctive peaks at 3335 and 2880 cm^{-1}, attributed to −OH and −CH stretching bands, respectively. After treatment with PEDOT:PSS, the broad peak at 3335 cm^{-1} disappeared, which might be due the cross-linking reactions between −OH groups of cellulose and the functionalized group of PEDOT:PSS.[37] Both the composite sample exhibit vibrational bands at 1343, 1148], and 1100 cm^{-1} that corresponds to C−C stretching band of the quininoid structure of the thiophene ring, the S−O bond in PSS, and the −CO band from the ether linkage of PEG that was codoped with PEDOT:PSS, respectively.[53] The peak observed at 1054 cm^{-1} is responsible for the S−phenyl bond in PSS. Additionally, bands at about 960 and 840 cm^{-1} originated from C−S stretching in the thiophene ring.[54]

Pristine cotton is an electrically insulating material; however, electrical conductivity can be introduced by incorporating nanomaterials such as PEDOT:PSS into its structure. To investigate the effect of PEDOT:PSS on the electrical conductivity of composite fabrics, the fabric was first dipped into the PEDOT:PSS solution, and subsequent dipping cycles were executed. As such, after 10 dipping cycles, the obtained sheet resistance was 2.52 kΩ/sq, whereas further increasing the number of dipping cycles to 20 led to a decrease in the sheet resistance to 1.05 kΩ/sq. Furthermore, the enhancement of the electrical conductivity can be realized from the codoping effect of PEG with PEDOT:PSS, where PEG supports the phase segregation between PEDOT−PSS-conducting particles and the excessively insulating PSSNa shell. The successful codoing effect is evidenced from the FTIR analysis (Figure 2b). In addition, it is assumed that the associated hydrogen

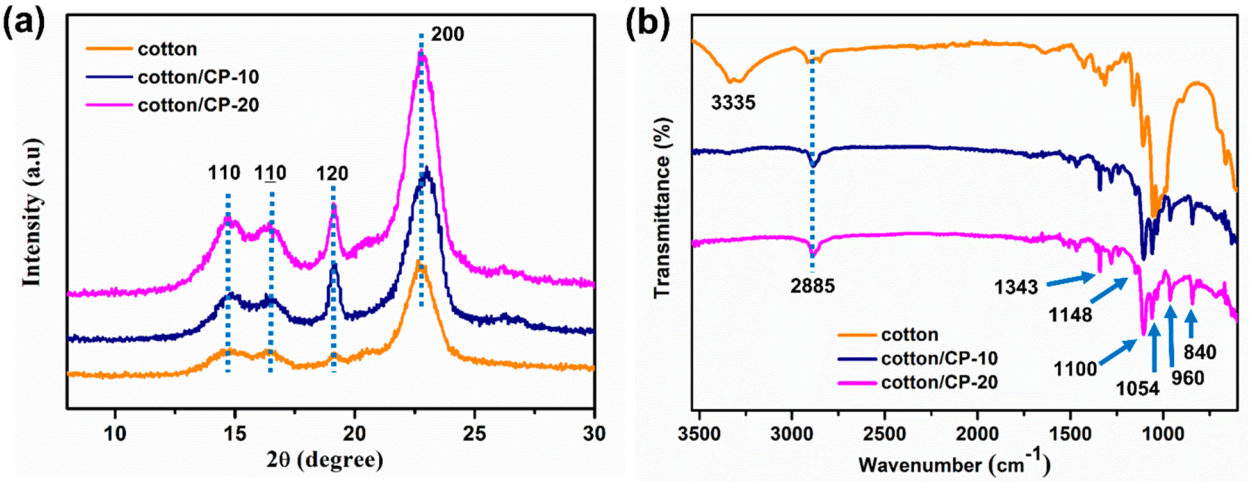

Figure 2. Structural characterizations of the cotton/CP heater. (a) Corresponding XRD patterns and (b) FTIR spectra of the samples.

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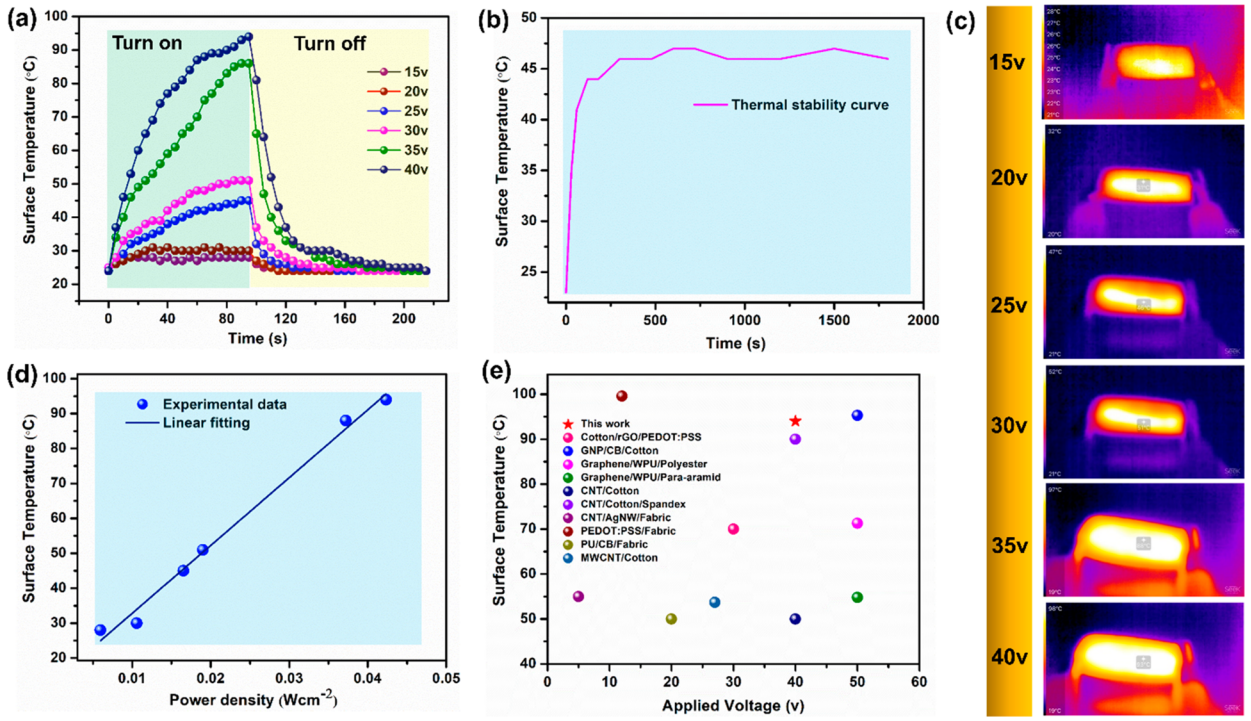

Figure 3. Joule heating characterization of the cotton/CP heater. (a) Time-dependent temperature profile of the heater against various voltages. (b) Thermal stability curve of the heater showing a stable heating performance for 30 min. The heater was evaluated at 25 V. (c) Thermal camera images of the samples under various voltages. (d) Plot of surface temperature versus the power density. (e) Comparison of the performance of the fabricated cotton/CP heater with those of others reported in the literature.[37,46,56−61]

bonds between PEG and PSSH weaken the electrostatic interaction between PEDOT and PSS.[55] Overall, these interactions afforded excellent conduction pathways and decreased the surface resistance of the fabric.

4.2. Joule Heating Performance of Cotton/CP Fabrics. The electrothermal behavior of cotton textiles coated with PEDOT:PSS under 20 dip-coating cycles (cotton/CP-20) was evaluated. The fabric with a size of 6 × 6 cm^2 was connected to a direct current (DC) supply, and various voltages (15−40 V) were applied. The surface temperature distribution morphology was recorded with a thermal IR camera.

The Joule heating characteristics can be observed from the inelastic collision between electrons and photons when the electrons pass through the materials in an electric field.[37] The time-dependent temperature profiles of the cotton/CP fabrics were categorized into three phases: heating state, steady-state, and cooling state. Figure 3a shows the time-dependent temperature profile of cotton/CP under different voltage supplies, revealing that the temperature gradually increases as the operating voltage and time increase. At the initial phase, i.e., the heating state, all cotton/CP samples illustrated an immediate increase in surface temperature at the applied voltage. Within 60 s, the surface temperature of the sample reached 26, 31, 40, 45, and 81 °C at the application voltages of 15, 20, 25, 30, and 35 V, respectively. However, an additional 40 s was required to achieve the steady-state temperature. The maximum heating temperature achieved for these samples varied from 26 to 94 °C when applied voltage varied from 15 to 40 V, respectively. The cooling state was largely influenced by the highest temperature raised.

The influence of strained and unstrained conditions on the heating performance was investigated. The time-dependent temperature profile at the unstrained condition is shown in Figure 3a. The sample operated at 25 V reached a steady-state temperature of 44 °C after 90 s, while the immediate release of the input voltage caused the sample to reach room temperature within only 24 s. Upon applying maximum driven voltages of 30 and 35 V, the surface temperature of the samples reached maxima of 51 and 70 °C, and the recovery time was calculated to 35 and 60 s, respectively. Similarly, the sample operated at 40 V required a similar time (∼90 s) to acquire a maximum surface temperature of 94 °C and then cooled to room temperature within 80 s.

The long-term heating stability of the Joule heater was examined at a convenient driving voltage. It is important to note that the driving voltage should be within the compatible range of the human body.[37] Therefore, 25 V was considered as the optimum driving voltage and was employed in further electrothermal studies. At 25 V, reasonable thermal stability was achieved as the heater exhibited a reliable heating performance of 1800 s, with a slight (4%) deterioration of heating performance (Figure 3b). In practice, these heating stabilities are highly desirable during the long-term and reliable operation of the heater.

Besides, the IR camera images revealed the uniform temperature morphology of the cotton/CP samples, Figure 3c. Thus, the Joule heating experimental results supported the morphological and chemical analyses of cotton fabric treated with PEDOT:PSS. This could be due to the effective functionalization and electroconductivity of cotton textiles, which consequently help to achieve a superior electrothermal heating performance. Moreover, Figure 3d suggests an almost linear increase in the surface temperature as a function of the input power, showing low power consumption from ∼0.005 to ∼0.04 W cm^{-2} that corresponds the temperature range from 26 to 94 °C. Figure 3e compares the Joule heating

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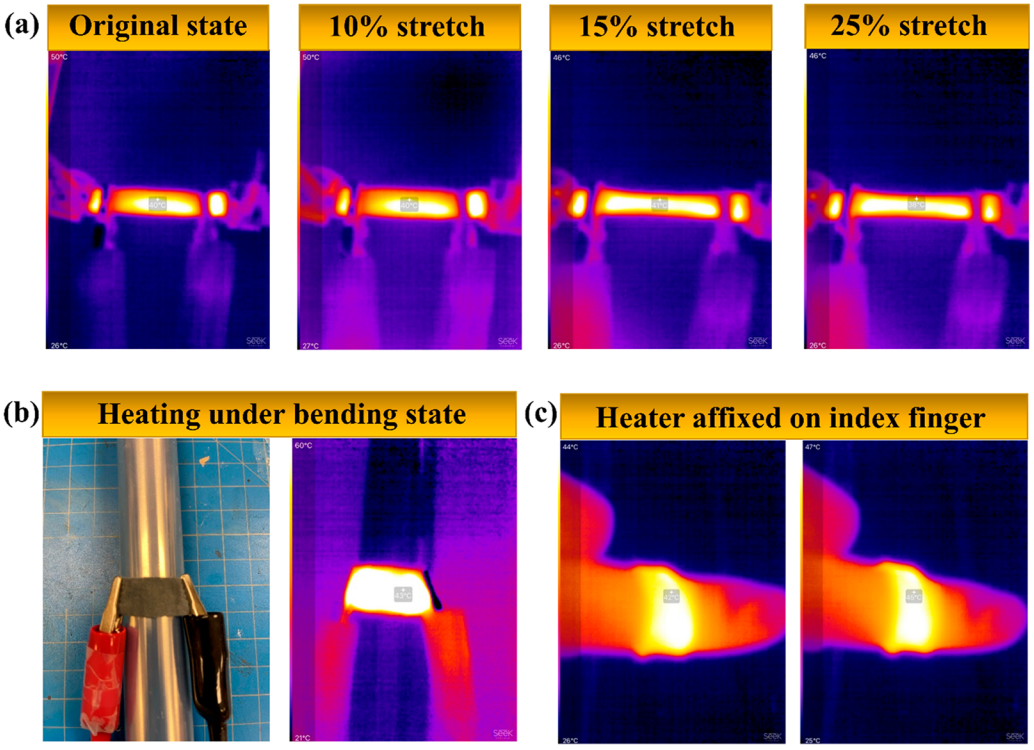

Figure 4. Evaluation of mechanical robustness and practical utilization of the cotton/CP heater. (a) Surface temperature morphology of the cotton/CP sample under various strained conditions. (b) Heating morphology of the prepared Joule heater under the bending state. (c) Image of the developed wearable heater attached on index finger, showing its uniform heating performance.

performance of various electrical heaters with the sample of the present investigation, showing the superiority of the performance of this sample over several heaters reported in the literature.

The heater should possess sufficient flexibility and mechanical robustness for wearable applications, as it might undergo several body deformations during application. In this sense, the Joule heating behavior of the heater was also evaluated at various strained and bending conditions. At the original state, the surface temperature of the heater was 40 °C, while at the subsequent 10% and 15% stretching conditions the heater exhibited no changes in the temperature profile. Consequently, when a considerable stretching condition of 25% was applied, the heater illustrated a negligible 2.5% decrease in the surface temperature. These could be due to the sliding of interstitches of the fabrics during stretching, which increases the sheet resistance. However, even after these activities, the surface temperature morphology of the stretched sample was found to be uniformly distributed (Figure 4a). Furthermore, to examine the heater's performance during bending phenomena, it was curved around a round-shaped tube. In this case, an excellent heating morphology was achieved and was distributed equally over the surface of the bent sample (Figure 4b). Finally, the heater was affixed on the index finger of a volunteer's hand for a conceptual wearable demonstration. Upon applying voltage, the heater efficiently warmed the designated area of the finger, and an excellent heating morphology was obtained (Figure 4c). The required temperature could easily be achieved by tuning the applied voltage based on the intended need of usage. Ideally, this exhibits the heater's huge potential avenues in wearable thermal therapy applications. Additionally, with its excellent heating capability and thermal stability, the fabricated heater may be regard as a strong candidate in defrosting, deicing, and demisting applications.

5. CONCLUSION

In this study, a scalable and facile fabrication route has been utilized to develop a flexible composite-fabric-based Joule heater by treating a cotton fabric with an intrinsically conducting polymer PEDOT:PSS. The fabricated material was characterized with various techniques to elucidate its structural and morphological attributes. The synergies obtained between the cotton and the conducting polymer were beneficial in the design of fabric-based Joule heating devices. The Joule heating experiments depicted superior electrothermal behaviors coupled with high surface temperature, excellent heating stability, and reliable heating performance over various mechanical deformations. The practical demonstration of the heater suggests its excellent potential in wearable thermal therapy. Further, such fabric-based Joule heating devices may also find applications in deicing, defrosting, window defogging.

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Notes

The authors declare no competing financial interest.

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