




A resistive ink based all-printed fabric heater integrated wearable thermotherapy device

Pramod Sankara Pillai¹, B. S. Athira^{2,3}, Harris Varghese^{2,3}, Shilpi Agarwal^{1,4}, Bipin Kumar¹, Ramasamy Alagirusamy¹, Apurba Das¹, Kuzhichalil Peethambaran Surendran^{2,3}, and Achu Chandran^{2,3,*} 

¹Department of Textile and Fibre Engineering, Laser Applications and Holography Laboratory, Instrument Design Development Centre, Indian Institute of Technology Delhi, New Delhi 110016, India

²Materials Science and Technology Division, CSIR-National Institute for Interdisciplinary Science and Technology (NIIST), Thiruvananthapuram 695019, India

³Academy of Scientific and Innovative Research (AcSIR), Ghaziabad 201002, India

⁴School of Physical Sciences, Jawaharlal Nehru University, New Delhi 110067, India

Received: 30 November 2022

Accepted: 16 May 2023

Published online:

1 June 2023

© The Author(s), under exclusive licence to Springer Science+Business Media, LLC, part of Springer Nature 2023

ABSTRACT

Owing to the current trends in the miniaturization of electronics, flexible and wearable gadgets are in great demand in various applications including therapeutic purposes. Fabric or textile-based devices are more preferred, as it blends with the outfit more than any other additional contraptions. Herein, an all-printed flexible fabric-based heater is developed. A silver-carbon composite ink is prepared to print on a polyester substrate in a meander configuration. The facile and low-cost heater fabricated via screen printing technology simply achieves a steady state temperature around 50 °C under 55 mW/cm² of applied electric power. Additionally, it exhibited a highly stable electro-thermal performance and uniform distribution of temperature over the whole area of the printed heater. The printed fabric heater is integrated into a thermotherapy wrist gadget for the purpose of superficial heat therapy. The ease in tunability of the temperature, conformity of the device to the skin, and the simplicity in fabrication enable the printed fabric heater integrated wearable band to be an excellent candidate for household thermotherapy applications.

1 Introduction

Thermotherapy, or heat therapy, is the use of heat for therapeutic purposes such as the treatment of musculoskeletal injuries [1]. It has widely proven to be

exceptional, especially for those related to muscle tension or spasm. With the application of heat to the skin, the blood vessels in the area widen and result in increased blood flow in the region. This in turn results in the relaxation of superficial muscles, reduction in muscle spasms, increase in elasticity of

Address correspondence to E-mail: achuchandran@niist.res.in

connective tissues, and alleviation of stiffness in joints [2]. Mitigation of the pain owing to the application of heat results mainly from analgesia via the pain gate theory [3]. The therapeutic effect increases the extensibility of collagen tissues and decreases joint stiffness and results in pain relief. Heat therapy is a simple and safe method for solving back pain, arthritis, healing, etc. Throughout human history, even from earlier civilizations, various thermotherapy techniques are widely used, such as sunbath, fire therapy, hot needling, paraffin wax, fluidotherapy, and laser therapy. Furthermore, different kinds of superficial heating are practiced based on the needs. Conductive methods, such as heat pads or paraffin wax baths, convective heating such as steam room as well as the sauna, and radiative heating using infrared light are commonly utilized for applying heat to the skin [4]. Similarly, ultrasonic wave therapy and short wave diathermy are used for the application of heat into deep tissues [5]. Also, there is another classification on the basis of type of heat; dry heat therapy and moist heat therapy, where dry heat therapy makes use of sources such as heating packs and pads while steamed towels, hot baths, or moist heating packs are utilized for moist heating.

Conventional thermotherapy methods such as heat packs and wraps are heavy, bulky, and mechanically rigid. Rather than these methods, electro-resistive heating (or Joule heating) can achieve the same feat, with a smaller form factor and ease. Heat is generated according to Joule's law when an electric current passes through the heater. The heat generated per unit time (H) is given by Eq. (1)

$$H = I^2 R, \quad (1)$$

where R is the resistance and I is the current. So, for effective heating, a reasonable resistivity is necessary for the conductor material. The system achieves a steady state temperature by balancing the Joule effect as well as other heat losses and then the power dissipated is directly proportional to that of the temperature. Heat loss is mainly due to thermal conduction to the substrate, convection to the surrounding air, and radiation emitted from the hot surface. So, the surface temperature depends upon the parameters such as heat capacity, mass, convective heat transfer co-efficient, emissivity, and conductivity of the heater as well as the surroundings. Here, the major advantage is that the temperature can be controlled very precisely by adjusting the voltage

or current in the circuit [6]. Recently, lightweight, body conformable, breathable, stretchable, and flexible resistive heaters have gained great attention owing to their various applications including wearable thermotherapy patches [7, 8].

With the advent of the Internet of Things (IoT) [9], miniaturizations of electronic devices is progressing quite rapidly. With this, the popularity of flexible electronics, mainly electronic or smart textiles are growing swiftly, which mainly incorporates textiles exhibiting conductivity or is a part of an electronic or computational purpose. The potential for multifunctional thermal management in emerging electronic applications has significantly increased the development of heaters with rapid thermal response, homogeneous temperature distribution, and extremely stable performance without thermal deterioration via joule heating effect. Xie et al. developed a joule heater with screen printed flexible aqueous MXene/xanthan gum hybrid inks which could reach a highest steady state temperature of 130.8 °C under 4 V of driving voltage [10]. Wang et al. could firstly formulate aqueous silver nanowire ink and ink jet printed on flexible polyethylene terephthalate (PET). Developed heater in mesh grid structure with 90% transparency could generate a steady state temperature of 100 °C at 4 V. The printed heater showed a high thermal response in 3 s due to insulating nature of PET [11]. A nature inspired approach adopted by Zhao et al. was able to produce 90.2 °C under 2 V from radial carbon scaffolds carbonized at 1000 °C [12]. A room temperature curable conductive ink was developed by Pillai et al. using the multiwalled carbon nanotube (MWCNT)-PEDOT:PSS nanocomposite which exhibited a highly uniform temperature distribution with a peak temperature of 136 °C and an electrical power density of 0.137 W/cm². Here, the role of PEDOT:PSS as an effective binder, dispersant, and conductivity enhancer in MWCNT ink is described successfully [13]. Gupta et al. fabricated a large area flexible heater with uniform temperature distribution, under low power by spray coating silver nanoparticle with crackle precursors. Transparent heater on PET substrate could effectively achieve a uniform temperature of 100 °C under an applied voltage below 5 V with fast response less than 20 s [7]. For healthcare applications with heaters, it is important to maintain contact with the skin for effective heat transfer and stable heating performance, even under large strains due to human motion. Therefore, implementing

conductive textiles as a heating source for ther-motherapy purposes is ideal. This could be achieved in two ways, either by adding the electronics to the textiles or by manufacturing the yarn with desired features [14]. Many different methods are available to fabricate conductive textiles, such as incorporating conductive metallic fibers into yarns [15], coating textiles with conducting materials, or preparing conducting polymer fibers to use directly as textiles. However, using metal fibers in the yarn is not preferable since the textile will lose its flexibility, softness, and the feel that is known and very closely associated with fabrics [16].

Treating or rather coating the fabrics with different materials such as metals [17, 18], conducting polymers [19, 20], or carbonaceous materials [21, 22] facilitates to incorporate heating into textiles. Zhang et al. [23] reported a process to transform commercial textiles and threads into sewable and weavable heaters for local climate control and personal thermal management. They replaced the conventional Joule heating element by conducting polymer poly(3,4-ethylenedioxythiophene) to be used as a thermal glove. Choi et al. [24] fabricated a soft, thin, and stretchable heater using a composite of silver nanowires and a thermoplastic elastomer. They created an electronic band for long-term, continuous articular thermotherapy. Jang et al. [25] developed a heater that exhibits high heating performance at low voltage and high thermal response by facile kirigami patterning of a highly conductive aluminum paper. The heater was attached to the wrist and activated. It was characterized by examining blood flow rate per volume with a laser doppler blood perfusion imager. High-efficiency electrothermal graphene oxide-based fabric joule heaters were fabricated by Tian et al. with the steady state temperature 162.6 °C and the maximum heating rate of 8.4 °C/s under 10 V applied voltage [26]. The spray coating process was adopted here for forming a bilayer structure of inner graphene/polyurethane composite and outer graphene oxide layer on cotton fabric. A wearable electronic device for human motion monitoring, health tracking and thermotherapy was developed by Zhao et al. from silver nanowire/wrap yarn [27]. The developed device possesses good sensitivity, high conductivity, high tolerable strain ~ 200% and high heating temperature (25–100 °C) even at low operation voltage (2–6 V). A smart heating control system with a novel sandwich structure was introduced by Huang et al.

which exhibited high thermostability, thermal resistance (163.5 °C/W cm²) and temperature sensitivity (0.135% °C⁻¹) [28]. The structure consists silver nanofiber (Ag NF)/silk fabric/platinum nanofiber (Pt NF) in which Ag NF network film functioned as wearable heater and Pt NF network arrays as temperature sensor. Multifunctional wearable electronic device based on silver fractal dendrites (Ag FDs) conductive ink with low-cost transfer printing method was developed by Tian et al. [29] Ag FDs ink-38 wt% deposited on polystyrene-block-polyisoprene-block-polystyrene (SIS) thin films showed high electrical conductivity (4.86×10^5 S/m), high sensing property, low-voltage driving Joule heating performance (52.3 °C at 1 V) with good mechanical stability. By chemically depositing silver nanoparticles (AgNPs) on cotton fabric, Niu et al. [30] created electrically and mechanically robust e-textiles that demonstrated low sheet resistance of 0.26 Ω/sq, high conductivity of 233.4 S/cm, and high stability to various mechanical deformations like ultrasonication, bending, and machine washing. The reason for a waterproof and machine-washable electronic gadget is the interface between AgNPs and fabric that has been strengthened and modified by bioinspired polydopamine and a fluorine-containing chemical called 1H,1H,2H,2H-perfluorodecanethiol (PFDT), respectively. By printing silver nanoparticle ink on commercially available medical grade tape, then covering the tape with silica nanoparticles as an ink-absorbing layer and chloride ions as a chemical sintering agent, Moon et al. showed instant, customized, and on-demand heat pads for thermotherapy [31]. The effect of thermotherapy is objectively confirmed using electroencephalography after the printed heat pad was attached to the shoulder. A self-heating stretchable laser induced graphene developed by Yang et al. could generate a temperature around 60 °C under 10 V with CO₂ laser scribing process, drop casting etc. [32].

For the development of conductive textile for thermotherapy or any other purpose, an alternative opportunity can also be followed, which is the technique of printing with ink that is conductive. This straightforward technique facilitates the ability to print any customized patterns at ease with the conductive ink of the choice on fabrics for heating applications [13]. In the present work, a flexible, facile, and screen printed fabric heater is fabricated using a silver-carbon composite ink on a polyester

substrate. This highly power-efficient inexpensive heater, designed in a meander configuration, is able to attain and maintain a steady state temperature of 50 °C by consuming around 55 mW/cm² of power. With the added flexibility of tuning the temperature with respect to the applied potential, this heater could be used as an excellent tool for wearable localized therapeutic platforms.

2 Results and discussion

As discussed earlier, printing the desired pattern into textiles is a very convenient method for fabric-based heaters. The obvious thing to initially consider is the ink that should be utilized for the printing process. The ink should exhibit conductive property; however, the heater should have an appreciable amount of resistance to deliver the desired heat. Therefore, a composite ink of silver and carbon inks was prepared by stirring both inks in 3-part silver and 2-part carbon. This grants the required conductivity as well as the necessary resistance to the ink. The main advantage of screen printing is the easiness to have any desired pattern for the heating element. However, for thermotherapy application, good area coverage with sufficient temperature output is required. The heater was designed in a meander pattern (Fig. 1a), which facilitates the necessary resistance because of the length of the design and the area coverage owing to the design itself. The detailed fabrication process is explained in the experimental section and the photograph of the final device is given in Fig. 1b. The

resistance of the heating element is measured using a digital multimeter by connecting its probes to the connection pads on the heater. The obtained value of resistance of the heating element from the end-to-end printed pattern is 30.81 Ω .

Before testing the heater in the real world, its performance can be understood with the help of COM-SOL Multiphysics software. A model of the heater in the exact dimensions was made on the finite element analysis software with the help of the CAD tools available (Fig. 2a). The necessary physics such as electric currents and Joule heating were added to the model. The model considers the heat transfer through conduction, heat loss through natural convection, and radiation to the environment. Then the proper parameters of the ink were also added to make the simulation more accurate. The whole model has appropriately meshed and a stationary study was evaluated to obtain the simulated temperature output of the heater. When the heat transfer through conduction is equal to the heat loss through convection and radiation, the system achieves a steady state condition. The steady-state temperature output of the heater obtained for a 5 V supply is 60 °C (Fig. 2b), where the required temperature for therapeutic heating is well within this range [25]. Furthermore, the simulation shows an almost uniform heat distribution in the 5 × 5 cm² heater area of the proposed heater design. Additionally, different substrates were also tried in the simulation to see their effect of it on the temperature output (Fig. S1). Overall, the simulation shows that the designed heater is more than

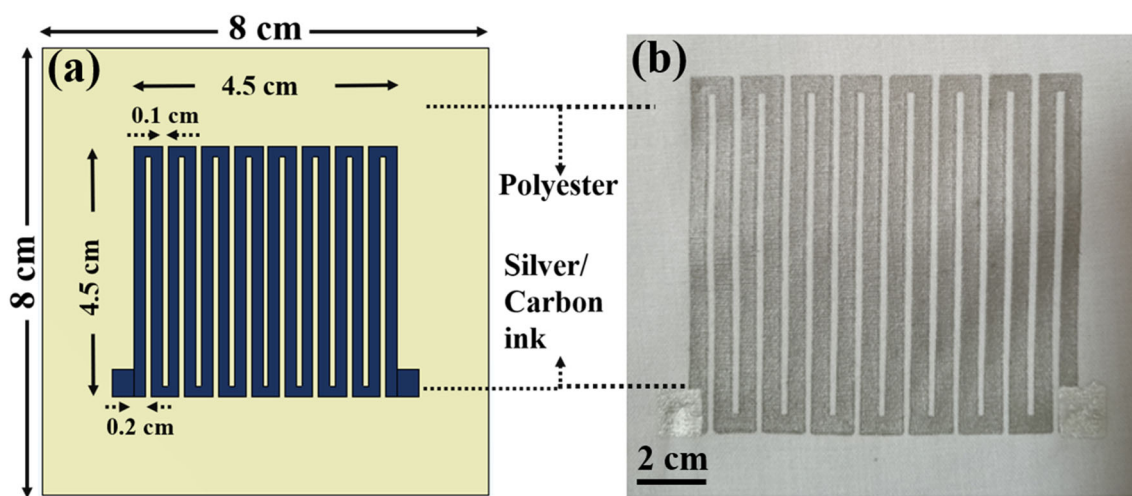


Fig. 1 **a** Schematic of meander-patterned heater and **b** the image of fabricated heater

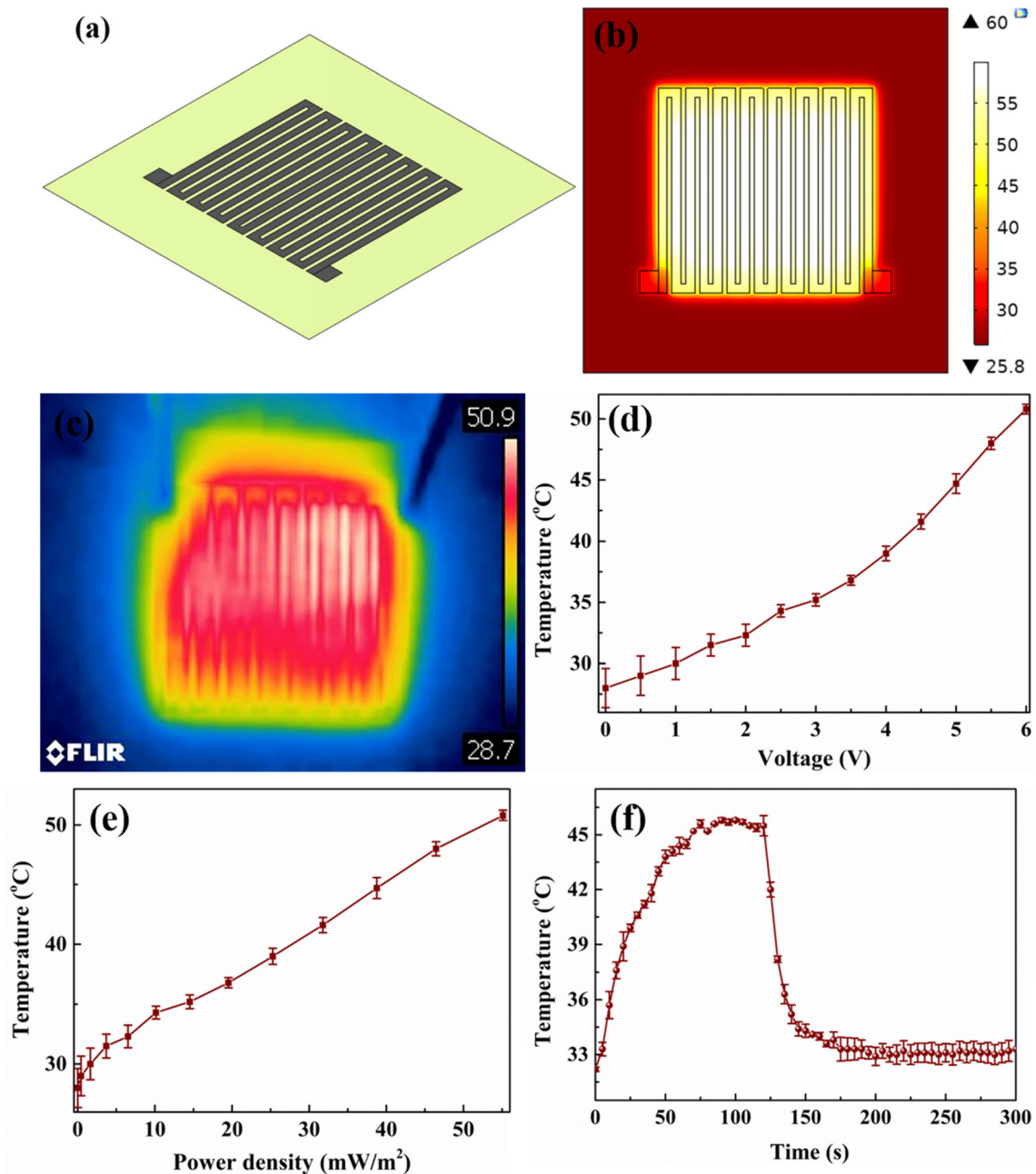


Fig. 2 Electro-thermal simulation using COMSOL Multiphysics. **a** Schematic of the model developed in COMSOL, and **b** temperature distribution of the heater at 5 V using COMSOL Multiphysics software. Electro-thermal characteristics of printed

fabric heater. **c** Temperature distribution at 5 V input bias, **d** temperature vs voltage, **e** temperature vs power, and **f** ON–OFF characteristics of the flexible fabric heater

capable of delivering the required temperature for thermotherapy.

Afterward, the all-printed fabric heater fabricated was tested for evaluating its electro-thermal performance. The characterization was done by noting the temperature by varying voltage from 0 to 5 V which is illustrated in Fig. S2. There is a uniform

distribution of temperature throughout the whole heating area and, it is clearly evident from the thermogram that the electrodes are colder than the area of the carbon-silver composite ink. Thus, the device is highly useful for producing localized heating for heat treatments. The heater could achieve a temperature around 50 °C under 5 V (Fig. 2c). These results were

also compared by measuring temperature using a thermocouple-based temperature sensor also. The temperature of the heater under different input voltage and power is shown in Fig. 2d and e. It is evident from the graphs that precise control of the output temperature is possible due to the response of the heater distinctly even for the small difference in input voltage (0.5 V). The fabric heater maintains a 50 °C surface temperature by consuming a very low power of 55 mW/cm², which makes it a very promising candidate for satisfying the global need for low energy consumption systems. Similarly, Fig. 2f shows the 'ON-OFF' characteristics of the sample under 5 V. The fabric heater device is relatively fast enough to reach a steady state temperature within 60 s on its 'ON' cycle and it reaches back to room temperature in and about another 60 s in its 'OFF' cycle.

As stated before, flexibility and conformity to the surface are the major advantages associated with an active fabric heater. A bending and twisting test were done for assessing the flexibility as well as stability of the heater. The results obtained are shown in Fig. 3. The heater was subtended on beakers of different radii of curvature (2.2 cm to 5.4 cm). Peak temperature at 5 V and resistance was measured in each condition and it was observed that there was no significant change in the temperature distribution as well as the resistance of the heater (Fig. 3a). This assures good flexibility to the heater, which could conform to the skin without affecting its performance. Similarly bending (Fig. 3b) and twisting tests

(Fig. 3c) were also done to showcase the stability of the fabricated heater. Neither the successive bending nor the twisting tests did affect the thermal performance of the fabricated heater.

To demonstrate the practical applicability of the meander heater as a wearable thermotherapy tool, the device was inserted into a band-aid and it was attached to the wrist as shown in Fig. 4a as a gadget. The temperature output for the same was measured as before and it was obtained to be 46 °C for a 5 V supply (Fig. 4b and supplementary video 1). Electro-thermal characteristics of the wearable device were obtained (Fig. 5a) for up to 5 V. It elucidates the fact that even with the additional thermal load of the band-aid, the temperature of the thermotherapy gadget can be tuned precisely. In a similar fashion, the temperature-power profile was also measured (Fig. 5b). 'ON-OFF' characteristics under 5 V were also done for this system which is shown in Fig. 5c. Due to the material property of band-aids, it took 150 s to attain steady state temperature at the surface, and similarly, the duration of cooling to room temperature also took a long time. The gadget increases the blood flow by inducing vasodilation which grants relief from pain and joint stiffness. Even though the heater is stable after bending and twisting, being cheap, it can be replaced easily if necessary. This method can also be extended to demonstrate various thermotherapy devices to cover various body parts. Moreover, the heater could be powered even by a 5 V power bank or using a mobile phone charger, and this solution is not bulky or inconvenient like the

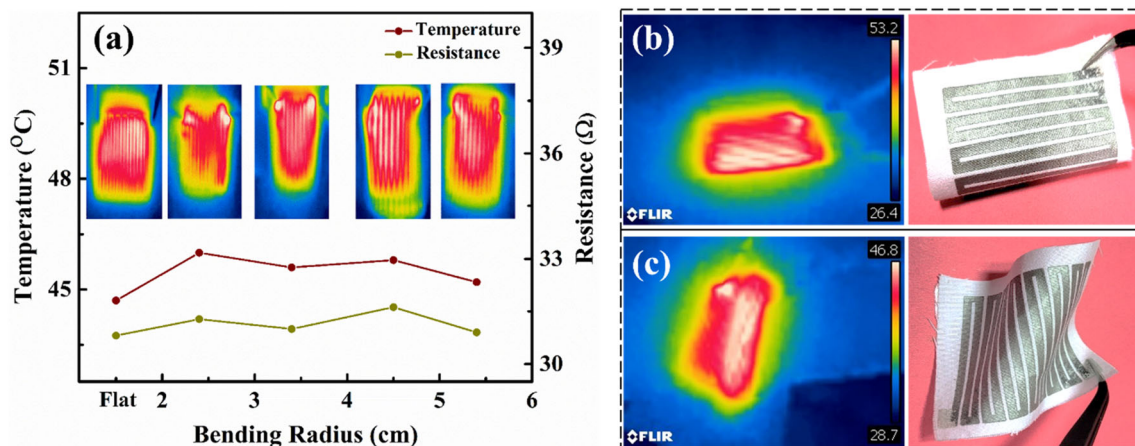


Fig. 3 Stability analysis of the heater. **a** Thermal images of the heater under different bending radii with its resistance as well as the temperature output of the same with completely, **b** bent and **c** twisted configurations

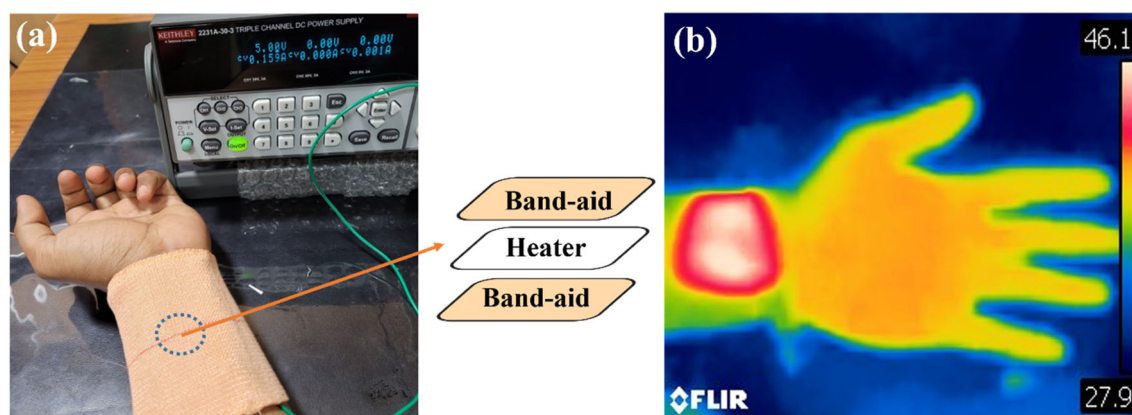


Fig. 4 **a** Photograph of a band-aid with integrated heater, **b** thermal image with temp. distribution at 5 V DC input

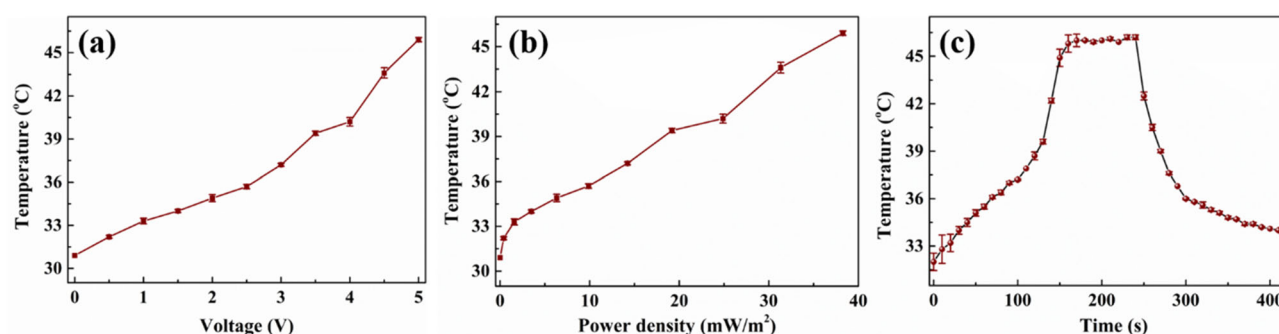


Fig. 5 Characterization of the band-aid with heater insert. **a** Temperature vs voltage, **b** temperature vs power density, and **c** ON–OFF Characteristics

present household thermotherapy solutions. Instead of using portable power bank, integration of flexible supercapacitors which possess high capacitance, energy density even in mechanical deformation is also a sustainable solution. Owing to the low power consumption of our device, a long-term usage is also possible. Many of the reported high performance flexible supercapacitors retain its capacitance under 0–180° bending, continuous twisting and bending as well as in mild conditions [33].

3 Experimental section/methods

3.1 Materials and methods

Commercial screen printable inks of carbon (Metalon® HPR-59) and silver (Metalon® HPS-021LV) were procured from Novacentrix, USA. All the materials were used without any further purifications. Silver–CARBON composite ink was prepared by mixing silver with carbon at a weight ratio

3:2 and homogenous ink was obtained by stirring it for 24 h continuously. Method of screen printing, with a printer (XPRT2, Ekra, Bonnigheim, Germany), was adopted for the fabrication of the fabric heater in a meander type pattern. Subsequently, the prepared ink was printed on a polyester substrate. Here, the printing parameters were optimized by trials for achieving the desired resistance of the heating element and thereby the operating voltage and temperature. The printed pattern was dried at 120 °C for 2 h. The heater is as shown in the schematic (Fig. 1a) with an area of $5 \times 5 \text{ cm}^2$ and two conducting electrodes as contact pads which are printed using silver ink. The image of the fabricated heater is shown in Fig. 1b.

The printed heater was integrated into a band-aid, by sandwiching the same in-between two layers of band-aids. Connections were taken out from the wearable gadget for powering the heater. The whole configuration can be easily wound tightly around limbs.

3.2 Characterization techniques

Printed heater was characterized for its electro-thermal performance and flexibility. Resistance of the heater was measured using Keithley DMM6500 6 ½ Digit Multimeter. The electro-thermal properties of the meander-patterned heater inserted band-aid were studied using Keithley 2231A-30-3 Triple Channel DC Power Supply and an IR imaging camera (C2 Compact Thermal Camera, FLIR, Wilsonville, USA).

4 Conclusions

The demonstrated meander-patterned flexible fabric heater can be efficiently used for therapeutic purposes. The facile and wearable fabric heater based on carbon-silver composite ink is able to sustain a stable temperature of about 50 °C for 55 mW/cm² of power. Moreover, it displayed excellent electro-thermal performance and the temperature is quite uniform throughout the whole surface of the 5 × 5 cm² heater. The heater showed no degradation in performance despite bending and twisting. This showcase the fact that the screen printing technique is an excellent fabrication method to obtain high-performing wearable fabric heaters in the desired patterns and sizes. In conjunction with the flexibility of the Joule heating, the developed flexible heater is an ideal candidate for localized conformal surface heating of the skin. Highly effective low voltage local heating of the wrist is achieved by attaching the heater inserted band-aid gadget. It is efficient for thermotherapy by increasing blood flow in a controllable manner and the same can be implemented into any body part in any desired configuration. The simplicity and ease of the all-printed flexible fabric heater promise great possibilities rather than the bulky alternatives such as the heating packs.

Acknowledgements

The authors sincerely thank Director, IIT Delhi, and Director, CSIR-NIIST for their continuous encouragement and support.

Author contributions

PSP: Conceptualization, methodology, formal analysis and investigation. Writing—original draft preparation. BSA: Methodology, formal analysis and investigation. Writing—original draft preparation. HV: Methodology, formal analysis and investigation. Writing—original draft preparation. SA: Formal analysis and investigation. BK: Writing—review and editing, supervision. RA: writing—review and editing, supervision. AD: Conceptualization, writing—review and editing, supervision. KPS: Conceptualization, formal analysis and investigation. Writing—original draft preparation, writing—review and editing, supervision. AC: Conceptualization, methodology, formal analysis and investigation. writing—original draft preparation, writing—review and editing, resources, supervision.

Funding

The authors declare that no funds, grants, or other support were received during the preparation of this manuscript.

Data availability

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

Declarations

Competing interests The authors have no relevant financial or non-financial interests to disclose.

Supplementary Information: The online version contains supplementary material available at <http://doi.org/10.1007/s10854-023-10665-7>.

References

1. L.M. Bearne, M.V. Hurley, *Rheumatology* (Elsevier, Amsterdam, 2010), pp.111–122
2. G.A. Malanga, N. Yan, J. Stark, *Postgrad. Med.* **127**, 57 (2015)
3. R. Melzack, P.D. Wall, *Science* **150**, 971 (1965)

4. L.H. Gerber, J.E. Hicks, *Osteoarthritis, Diagnosis and Medical/Surgical Management*, 2nd edn. (W.B. Saunders Company, Philadelphia, 1992), pp.427–464
5. H. Daga, S. Raval, H. Toshikhane, N. Jethava, *World J. Pharm. Res.* **6**, 1602 (2019)
6. D.T. Papanastasiou, A. Schultheiss, D. Muñoz-Rojas, C. Celle, A. Carella, J. Simonato, D. Bellet, *Adv. Funct. Mater.* **30**, 1910225 (2020)
7. R. Gupta, K.D.M. Rao, S. Kiruthika, G.U. Kulkarni, *ACS Appl. Mater. Interfaces* **8**, 12559 (2016)
8. Z. Yu, Y. Gao, X. Di, H. Luo, *RSC Adv.* **6**, 67771 (2016)
9. M.H. Miraz, M. Ali, P.S. Excell, R. Picking, *2015 Internet Technologies and Applications (ITA)* (IEEE, 2015), pp. 219–224.
10. H. Wu, Y. Xie, Y. Ma, B. Zhang, B. Xia, P. Zhang, W. Qian, D. He, X. Zhang, B.-W. Li, C.-W. Nan, *Small* **18**(16), 2107087 (2022)
11. P.-H. Wang, S.-P. Chen, C.-H. Sua, Y.-C. Liao, *RSC Adv.* **5**, 98412–98418 (2015)
12. B. Zhao, P. Bai, S. Wang, H. Ji, B. Fan, R. Zhang, R. Che, *ACS Appl. Mater. Interfaces* **13**(24), 29101–29112 (2021)
13. A.S. Pillai, A. Chandran, S.K. Peethambharan, *Appl. Mater. Today* **23**, 100987 (2021)
14. O. Kayacan, E. YazganBulgun, *Int. J. Cloth. Sci. Technol.* **21**, 127 (2009)
15. K. Chatterjee, J. Tabor, T.K. Ghosh, *Fibers* **7**, 51 (2019)
16. A. Schwarz, I. Kazani, L. Cuny, C. Hertleer, F. Ghekiere, G. De Clercq, L. Van Langenhove, *Text. Res. J.* **81**, 1713 (2011)
17. S. Hong, H. Lee, J. Lee, J. Kwon, S. Han, Y.D. Suh, H. Cho, J. Shin, J. Yeo, S.H. Ko, *Adv. Mater.* **27**, 4744 (2015)
18. H. Liu, J. Li, L. Chen, L. Liu, Y. Li, X. Li, X. Li, H. Yang, *Text. Res. J.* **86**, 1398 (2016)
19. Q.-W. Wang, H.-B. Zhang, J. Liu, S. Zhao, X. Xie, L. Liu, R. Yang, N. Koratkar, Z.-Z. Yu, *Adv. Funct. Mater.* **29**, 1806819 (2019)
20. M.R. Moraes, A.C. Alves, F. Toptan, M.S. Martins, E.M.F. Vieira, A.J. Paleo, A.P. Souto, W.L.F. Santos, M.F. Esteves, A. Zille, *J. Mater. Chem. C Mater.* **5**, 3807 (2017)
21. A. Levitt, J. Zhang, G. Dion, Y. Gogotsi, J.M. Razal, *Adv. Funct. Mater.* **30**, 2000739 (2020)
22. Y. Liu, X. Wang, K. Qi, J.H. Xin, *J. Mater. Chem.* **18**, 3454 (2008)
23. L. Zhang, M. Baima, T.L. Andrew, *ACS Appl. Mater. Interfaces* **9**, 32299 (2017)
24. S. Choi, J. Park, W. Hyun, J. Kim, J. Kim, Y.B. Lee, C. Song, H.J. Hwang, J.H. Kim, T. Hyeon, D.-H. Kim, *ACS Nano* **9**, 6626 (2015)
25. N.-S. Jang, K.-H. Kim, S.-H. Ha, S.-H. Jung, H.M. Lee, J.-M. Kim, *ACS Appl. Mater. Interfaces* **9**, 19612 (2017)
26. M. Tian, Y. Hao, L. Qu, S. Zhu, X. Zhang, S. Chen, *Mater. Lett.* **234**, 101 (2019)
27. M. Zhao, D. Li, J. Huang, D. Wang, A. Mensah, Q. Wei, J. Mater. Chem. C Mater. **7**, 13468 (2019)
28. J. Huang, Y. Li, Z. Xu, W. Li, B. Xu, H. Meng, X. Liu, W. Guo, *Nanotechnology* **30**, 325203 (2019)
29. B. Tian, Q. Liu, C. Luo, Y. Feng, W. Wu, *Adv. Electron. Mater.* **6**(2), 2070006 (2020)
30. B. Niu, S. Yang, T. Hua, X. Tian, M. Koo, *Nano Res.* **14**, 1043 (2021)
31. D. Moon, G. Plečkaitytė, T. Choi, M. Seol, B. Kim, D. Lee, J. Han, M. Meyyappan, *Adv. Healthc. Mater.* **9**, 1 (2020)
32. Li. Yang, N. Yi, J. Zhu, Z. Cheng, X. Yin, X. Zhang, H. Zhu, H. Chen, *J. Mater. Chem. A* **8**, 6487–6500 (2020)
33. Z. Zhao, K. Xia, Y. Hou, Q. Zhang, Z. Ye, J. Lu, *Chem. Soc. Rev.* **50**(22), 12702 (2021)

Publisher's Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Springer Nature or its licensor (e.g. a society or other partner) holds exclusive rights to this article under a publishing agreement with the author(s) or other rightsholder(s); author self-archiving of the accepted manuscript version of this article is solely governed by the terms of such publishing agreement and applicable law.

Terms and Conditions

Springer Nature journal content, brought to you courtesy of Springer Nature Customer Service Center GmbH (“Springer Nature”).

Springer Nature supports a reasonable amount of sharing of research papers by authors, subscribers and authorised users (“Users”), for small-scale personal, non-commercial use provided that all copyright, trade and service marks and other proprietary notices are maintained. By accessing, sharing, receiving or otherwise using the Springer Nature journal content you agree to these terms of use (“Terms”). For these purposes, Springer Nature considers academic use (by researchers and students) to be non-commercial.

These Terms are supplementary and will apply in addition to any applicable website terms and conditions, a relevant site licence or a personal subscription. These Terms will prevail over any conflict or ambiguity with regards to the relevant terms, a site licence or a personal subscription (to the extent of the conflict or ambiguity only). For Creative Commons-licensed articles, the terms of the Creative Commons license used will apply.

We collect and use personal data to provide access to the Springer Nature journal content. We may also use these personal data internally within ResearchGate and Springer Nature and as agreed share it, in an anonymised way, for purposes of tracking, analysis and reporting. We will not otherwise disclose your personal data outside the ResearchGate or the Springer Nature group of companies unless we have your permission as detailed in the Privacy Policy.

While Users may use the Springer Nature journal content for small scale, personal non-commercial use, it is important to note that Users may not:

1. use such content for the purpose of providing other users with access on a regular or large scale basis or as a means to circumvent access control;
2. use such content where to do so would be considered a criminal or statutory offence in any jurisdiction, or gives rise to civil liability, or is otherwise unlawful;
3. falsely or misleadingly imply or suggest endorsement, approval, sponsorship, or association unless explicitly agreed to by Springer Nature in writing;
4. use bots or other automated methods to access the content or redirect messages
5. override any security feature or exclusionary protocol; or
6. share the content in order to create substitute for Springer Nature products or services or a systematic database of Springer Nature journal content.

In line with the restriction against commercial use, Springer Nature does not permit the creation of a product or service that creates revenue, royalties, rent or income from our content or its inclusion as part of a paid for service or for other commercial gain. Springer Nature journal content cannot be used for inter-library loans and librarians may not upload Springer Nature journal content on a large scale into their, or any other, institutional repository.

These terms of use are reviewed regularly and may be amended at any time. Springer Nature is not obligated to publish any information or content on this website and may remove it or features or functionality at our sole discretion, at any time with or without notice. Springer Nature may revoke this licence to you at any time and remove access to any copies of the Springer Nature journal content which have been saved.

To the fullest extent permitted by law, Springer Nature makes no warranties, representations or guarantees to Users, either express or implied with respect to the Springer nature journal content and all parties disclaim and waive any implied warranties or warranties imposed by law, including merchantability or fitness for any particular purpose.

Please note that these rights do not automatically extend to content, data or other material published by Springer Nature that may be licensed from third parties.

If you would like to use or distribute our Springer Nature journal content to a wider audience or on a regular basis or in any other manner not expressly permitted by these Terms, please contact Springer Nature at

onlineservice@springernature.com