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LETTER

Low-cost and customizable inkjet printing for microelectrodes fabrication

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Abstract

Microelectrodes for detection of chemicals present several advantages over conventional sized electrodes. However, rapid and low-cost fabrication of microelectrodes is challenging due to high complexity of patterning equipment. We present the development of a low-cost, customizable inkjet printer for printing nanomaterials including carbon nanotubes for the fabrication of microelectrodes. The achieved spatial resolution of the inkjet printer is less than 20 µm, which is comparable to advanced commercially available inkjet printers, with the advantage of being low-cost and easily replicated.

Keywords: Inkjet printing, Carbon nanotubes, Microelectrodes

Introduction

Inkjet printing has seen tremendous development in the deposition of nanomaterials for the fabrication of microelectrodes. It has been considered as an alternative for printing nanomaterials in the applications of flexible electronics and point of care sensors. The superior properties of inkjet printing such as solution processing and maskless printing enable the technology to be used in a variety of applications. A common issue in the development of inkjet-printed sensors is the high cost of inkjet printing research equipment.

Classically used for printing text and images, inkjet printing has recently found applications in many branches. Its advantages include the ability to print solutions of various materials, the extremely low waste and fine control of deposition parameters such as droplet location and number of printed droplets. Inkjet printing has been used to print thin film transistors on plastic substrates [1], fabricate stretchable FET transistors [2], tactile sensors [3], two dimensional force sensors [4], biosensors [5], chemiresistive sensors [6] and a variety of other applications [7]. While many of the above-mentioned studies used professional inkjet printers, some employed office inkjet printers which are widely available at low cost. Office inkjet printers offer the advantage of printing large area patterns with high speed, at the expense of lower resolution and lack of control of the quantity of material deposited.

Several approaches have been employed in the development of low-cost ink deposition processes. In microfluidic impact printing (MIP) [8, 9] a pin which is driven by an electromagnetic actuator strikes an elastic membrane. The ink contained in the microfluidic channel below the membrane ejects in the form of a small droplet. In electrohydrodynamic jet printing [10] a high voltage is applied between the print head and the substrate, which ejects ink from the nozzle onto the moving substrate to form a pattern. Printed lines with widths as small as 700 nm can be achieved. These approaches provide an alternative to standard printing procedures, however they are not easily replicated since the cartridge and nozzle have to be microfabricated in both cases.

Here, we present a customized inkjet printer with comparable characteristics to advanced commercially available inkjet printers, while being affordable by utilizing off-the-shelf components. Furthermore, it is capable of printing several nanomaterial solutions including carbon nanotubes, graphene oxide, and silver nanoparticles. We

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demonstrate the capabilities of the printer by patterning carbon nanotubes on polymer substrates. This inkjet printer can easily be replicated and provides a foundation for development of flexible and disposable sensors.

Experimental methods

Development of the inkjet printer stage

The inkjet printer consists of a moving stage, a print head with cartridge, and a control board. An aluminum frame was used as the platform for the inkjet printer. The frame offers a large area of 35 cm × 35 cm for positioning the substrate as illustrated in Fig. 1a. The control board contains the microcontroller and two subsystems: a motor control for positioning of the print head and a cartridge control that controls the ejection of droplets (Fig. 1b). Stepper motors with a geared system providing 2060 steps per revolution were used for X and Y linear movements of the cartridge. Inkjet printer cartridges (HP C6602A) were fully cleaned in deionized water and dried before use. The cartridge has a nozzle with aperture of 55 µm (measured under optical microscope) with a nominal average drop volume of 160 pl (obtained from the datasheet) [11]. Each droplet is generated by an electric pulse of 21 V for 5 µs sent by the microcontroller to the nozzle (Fig. 1c). This type of cartridge uses the thermal technology to generate droplets. When a voltage pulse is sent to the nozzle, a thin film resistor in the nozzle heats to a high temperature, creating bubbles and ejecting the ink [7]. Line patterns with width as low as 120 µm and square patterns were printed on poly(ethylene terephthalate) (PET) substrate to demonstrate the characteristics of the printer (Fig. 1d).

Ink solution preparation

Ink development is an important step for successful inkjet printing. For the nanomaterials utilized in the custom inkjet printer, a surfactant was added to provide a stable dispersion. The optimal formulation for inkjet printing on PET film has been previously developed in our group [12]. For the ink preparation, 10 mg/ml of

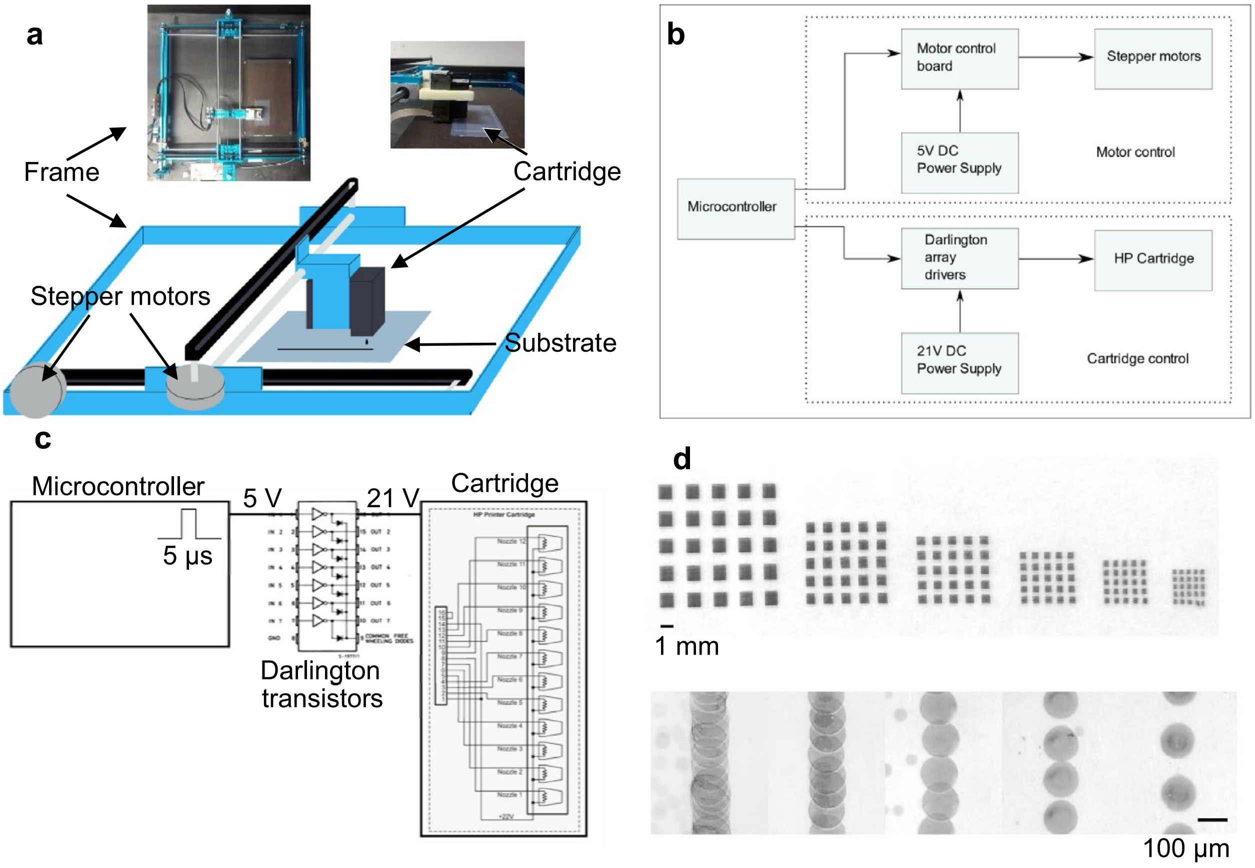

Fig. 1 System schematic and customized patterns: **a** XY stage containing the print head and cartridge; **b** schematic of the control board; **c** the microcontroller sends voltage pulses of 5 µs of duration and 21 V of amplitude to activate a particular nozzle, and **d** carbon nanotube patterns printed on a poly(ethylene terephthalate) (PET) film (scale bar: 1 mm, above) and a single line of carbon nanotubes with different spacing between drops (scale bar: 100 µm, below)

multi-walled carbon nanotube (MWCNT) (Cheap Tubes Inc., Brattleboro, VT, USA) and 7 mg/ml of sodium dodecyl sulfate (SDS) (Alfa Aesar, Ward Hill, MA, USA) were added to 5 ml of deionized water in a vial, sonicated for 30 min at 80 W (Fisher Scientific FS20D), transferred to centrifuge tubes and centrifuged for 5 min at 12,000 rpm. The supernatant was recovered and directly injected into a cartridge. The cartridge was opened and thoroughly cleaned before addition of the ink.

Printer characterization

For characterization of the inkjet printer, PET sheets (Inkpress ITF851150) were cut into 70 mm × 50 mm and used as the substrate. In order for the substrate to maintain flatness during printing, it was attached to a glass slide which was previously coated with poly(dimethyl siloxane) (PDMS). All measurements of dimensions were performed under a stereomicroscope (Leica MZ16), while sheet resistances were measured with a custom four-point probe setup. The current low-cost inkjet printer does not contain a heating stage, so the experiments were performed at room temperature. Alternatively, a heating stage would allow improvement of the evaporation rate of the droplets.

Results and discussion

Performance of the custom printer

Drop spacing

For performance testing of the developed printer, a single nozzle was used. The advantages are high spatial resolution and control of the number of droplets. The position of the cartridge is controlled by stepper motors. Each step of the motor corresponds to the minimum distance between drops printed on the substrate. Due to the reduction gears of the stepper motors, a single step corresponds to 18 µm, two steps correspond to 36 µm and so on. The spatial resolution of the inkjet printer is the smallest lateral displacement of the nozzle achievable for a single motor step, corresponding to a resolution of 18 µm (1412 dpi). In Fig. 2, lines of carbon nanotubes were printed on PET film to determine the drop spacing. The points represent the average for each number of steps, while the error bars represent the standard deviation of the distance between adjacent drops for 20 times of printing (n = 20). The deviations of the droplets both in the direction of the line and laterally were measured to obtain the positioning repeatability, achieving a standard deviation (SD) of 10.3 µm and 5.01 µm, respectively. The deviations can be attributed to the differences in trajectories of the ejecting droplets due to the distance between the nozzle and substrate.

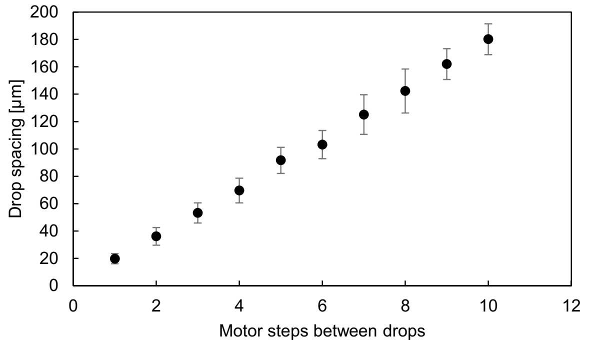

Fig. 2 Drop spacing measurement of the custom printer. Carbon nanotube lines were printed with each line having a different number of motor steps between drops. Each step of the motor, therefore, corresponds to 18 µm. Error bars show standard deviation of 20 measurements

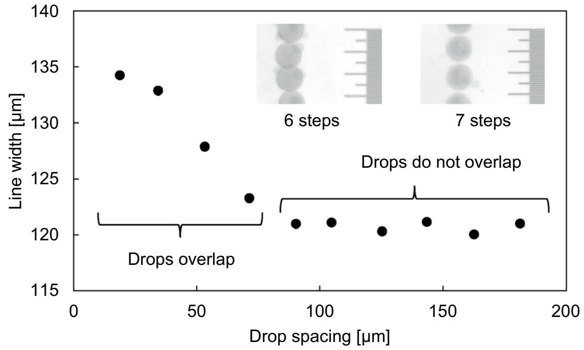

Fig. 3 Line width dependence on the drop spacing. For small drop spacing there is overlap and the line width increases

Line width

When printing a single line of nanomaterial on the substrate, adjacent drops in close proximity produce an increased line width. Figure 3 shows the dependence of line width on the drop spacing. If the spacing between each drop is less than 70 µm, drops may overlap. When drops overlap, the resultant line width increases. But if the drops do not overlap, the line width is the same as the drop diameter. Several methods can be used to change the size of the printed droplet, such as modifying the nozzle diameter, changing the type of substrate, the ink formulation and the hydrophilicity of the substrate to improve the wetting behavior [13]. In our experiments, changing the voltage and the pulse time did not affect the size of the printed droplet.

Sheet resistance

Conductivity of the printed patterns is important when fabricating functional sensors. As can be seen in Fig. 4, the measured sheet resistance is lower when drops are

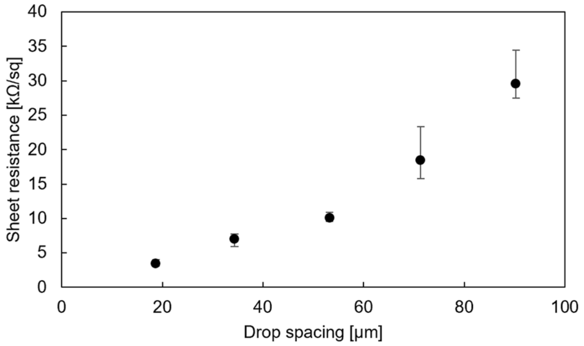

Fig. 4 Sheet resistance of single lines of carbon nanotubes printed on PET. As the drop spacing increases, less material is printed on given area and the sheet resistance increases. Error bars show (max-avg) and (avg-min) of 5 measurements

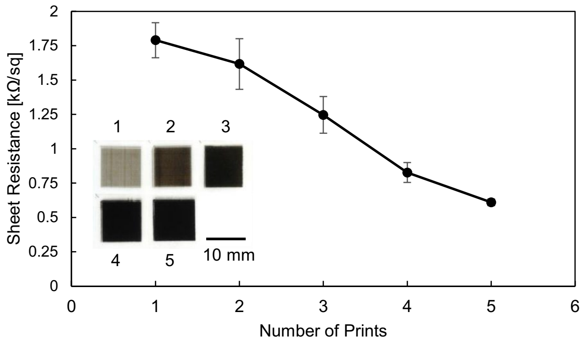

Fig. 5 Measured sheet resistance of carbon nanotubes on PET printed from one to five times. Error bars show standard deviation of measurements

printed closer together. The reason is obvious that, if the drops overlap, more material is printed in a given area, which in turn decreases the sheet resistance. On the contrary, as the drop spacing increases less material is printed on a given area and the sheet resistance increases. If the drop spacing is greater than 90 μm drops do not overlap and a conductive path is not formed.

In Fig. 5, square patterns of carbon nanotubes were printed on the PET sheet with dimensions of 10 mm × 10 mm. The measured sheet resistance decreases as a function of the number of prints due to more conduction paths available for current flow.

Table 1 shows the comparison of patterns deposited with a variety of inkjet printers and some representative performance parameters (sheet resistance, minimum line width).

Inkjet printing of humidity sensor

Humidity sensors have attracted interest due to their potential applications in indoor air quality and respiration detection [19, 20]. Several flexible and wearable sensors based on carbon nanotubes composites have been shown [21, 22]. To demonstrate the applications of the custom inkjet printer, a relative humidity (RH) sensor was fabricated by printing a microelectrode of carbon nanotubes on PET sheet and subsequently transferring the pattern to PDMS. A single line of carbon nanotubes was printed 30 times, corresponding to a line width of ~ 120 μm and length of 12 mm. The extremities were attached to copper wires through silver epoxy. The sensor was placed in an environment chamber with RH of 88% and subsequently removed from the chamber to ambient RH of 60%. As illustrated in Fig. 6, the resistance quickly rises upon change of the relative humidity, and returns to initial resistance when placed at ambient relative humidity. The response time achieved was similar to other humidity sensors based on carbon nanotubes [23]. The change is resistance was 2.6% when the humidity changed from 60 to 88%, or 0.093%/%RH.

Table 1 Comparison of inkjet printing performance

Inkjet printer	Printed material	Substrate	Sheet resistance/resistivity	Minimum line width	Ref
MicroJet	SWCNT	Glass	25 μΩ·m at 8 prints	150 μm	[14]
Canon BJC 4550	MWCNT	Transparency foil	40 kΩ/sq at 90 prints	200 μm	[15]
Micromech Systems	MWCNT	Glass	30 kΩ/sq at 1 print	98 μm	[16]
Microfab JetDrive III	MWCNT	PET film	0.7 kΩ/sq at 14 prints	100 μm	[17]
Dimatix DMP-8200	Ag nanoparticles	Paper	0.083 mΩ·m	59 μm	[18]
Custom (MIP)	Biological reagents	PDMS	NA	100 μm	[8]
Custom (EJP)	PEDOT/PSS, polyurethane prepolymer	Silicon wafer	NA	700 nm	[10]
Custom	MWCNT	PET film	0.61 kΩ/sq at 5 prints	120 μm	This work

NA Not available, *PDMS* poly(dimethyl siloxane), *EJP* electrohydrodynamic jet printing, *MIP* microfluidic impact printing, *SWCNT* single-walled carbon nanotube, *MWCNT* multi-walled carbon nanotube, *PEDOT/PSS* poly(3,4-ethylenedioxythiophene)/poly(styrenesulphonate)

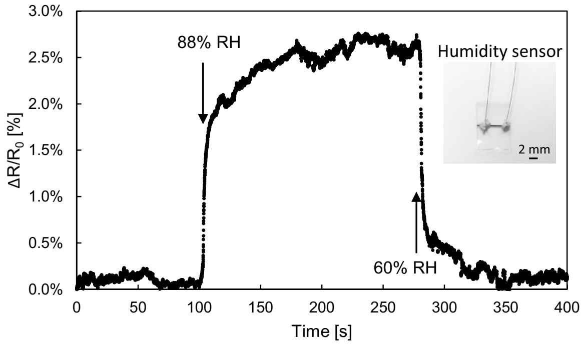

Fig. 6 Relative humidity sensor based on carbon nanotubes microelectrode printed with inkjet printer

Conclusions

We have developed a cost effective and customizable inkjet printer offering a resolution of 18 μm (1412 dpi) with the components available off-the-shelf, which is comparable to the advanced inkjet printers available for research. The inkjet printer was employed to print patterns of carbon nanotubes on PET film to demonstrate the capability of printing microelectrodes, which can be used in several applications such as electrochemical and humidity sensors. A relative humidity sensor was fabricated by printing carbon nanotube onto PET film and subsequently transferring to PDMS. The sensor showed 2.6% change in resistance when the humidity changed between 60 and 88% (from ambient to a high humidity environment). In addition, the flexibility of the sensor allows the integration in wearable applications for humidity monitoring.

Abbreviations

MIP: microfluidic impact printing; PET: poly(ethylene terephthalate); MWCNT: multi-walled carbon nanotube; SDS: sodium dodecyl sulfate; PDMS: poly(dimethyl siloxane); RH: relative humidity.

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Authors' contributions

THC and JWC designed the research. THC performed the experiments and drafted the manuscript. JWC supervised the research and drafting the manuscript. All authors read and approved the final manuscript.

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Availability of data and materials

All data generated or analyzed during this study are included in this published article.

Competing interests

The authors declare that they have no competing interests.

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References

1. Sajed F, Rutherglen C (2013) All-printed and transparent single walled carbon nanotube thin film transistor devices. Appl Phys Lett 103:143303. https://doi.org/10.1063/1.4824475
2. Molina-Lopez F, Gao TZ, Kraft U, Zhu C, Öhlund T, Pfattner R, Feig VR, Kim Y, Wang S, Yun Y, Bao Z (2019) Inkjet-printed stretchable and low voltage synaptic transistor array. Nat Commun 10:2676. https://doi.org/10.1038/s41467-019-10569-3
3. Salim A, Lim S (2017) Review of recent inkjet-printed capacitive tactile sensors. Sensors 17:2593. https://doi.org/10.3390/s17112593
4. da Costa TH, Choi J-W (2017) A flexible two dimensional force sensor using PDMS nanocomposite. Microelectron Eng 174:64–69. https://doi.org/10.1016/j.mee.2017.02.001
5. Gonzalez-Macia L, Morrin A, Smyth MR, Killard AJ (2010) Advanced printing and deposition methodologies for the fabrication of biosensors and biodevices. Analyst 135:845–867. https://doi.org/10.1039/B916888E
6. Song E, da Costa TH, Choi J-W (2017) A chemiresistive glucose sensor fabricated by inkjet printing. Microsyst Technol 23:3505–3511. https://doi.org/10.1007/s00542-016-3160-4
7. Tortorich R, Choi J-W (2013) Inkjet printing of carbon nanotubes. Nanomaterials 3:453–468. https://doi.org/10.3390/nano3030453
8. Ding Y, Huang E, Lam KS, Pan T (2013) Microfluidic impact printer with interchangeable cartridges for versatile non-contact multiplexed micropatterning. Lab Chip 13:1902. https://doi.org/10.1039/c3lc41372a
9. Mao Y, Wang X, Li X, Li B, Chu J (2019) Modelling on the droplet formation and optimizing of the microfluidic cartridge used for the microfluidic impact printing. J Micromech Microeng 29:125016. https://doi.org/10.1088/1361-6439/ab4cda
10. Park J-U, Hardy M, Kang SJ, Barton K, Adair K, Mukhopadhyay Kishore D, Lee CY, Strano MS, Alleyne AG, Georgiadis JG, Ferreira PM, Rogers JA (2007) High-resolution electrohydrodynamic jet printing. Nat Mater 6:782–789. https://doi.org/10.1038/nmat1974
11. Hewlett-Packard Company (2003) HP Extended TIJ 1.0 Print Cartridges. https://be02.cp-static.com/objects/pdf/3/3bf/31683_1_printer-en-scannerkits-hp-c6602a-black-c6602r-red-c6602g-green-c6602b-blue-96dpi-160pl-18cc-q2299a.pdf. Accessed 7 Nov 2019
12. Tortorich RP, Song E, Choi J-W (2014) Inkjet-printed carbon nanotube electrodes with low sheet resistance for electrochemical sensor applications. J Electrochem Soc 161:B3044–B3048. https://doi.org/10.1149/2.008402jes
13. Aleeva Y, Pignataro B (2014) Recent advances in upscalable wet methods and ink formulations for printed electronics. J Mater Chem C 2:6436–6453. https://doi.org/10.1039/C4TC00618F
14. Song J-W, Kim J, Yoon Y-H, Choi B-S, Kim J-H, Han C-S (2008) Inkjet printing of single-walled carbon nanotubes and electrical characterization of the line pattern. Nanotechnology 19:095702. https://doi.org/10.1088/0957-4484/19/9/095702
15. Kordás K, Mustonen T, Tóth G, Jantunen H, Lajunen M, Soldano C, Talapatra S, Kar S, Vajtai R, Ajayan PM (2006) Inkjet printing of electrically conductive patterns of carbon nanotubes. Small 2:1021–1025. https://doi.org/10.1002/smll.200600061
16. Wang T, Roberts MA, Kinloch IA, Derby B (2012) Inkjet printed carbon nanotube networks: the influence of drop spacing and drying on electrical properties. J Phys Appl Phys 45:315304. https://doi.org/10.1088/0022-3727/45/31/315304
17. Azoubel S, Shemesh S, Magdassi S (2012) Flexible electroluminescent device with inkjet-printed carbon nanotube electrodes. Nanotechnology 23:344003. https://doi.org/10.1088/0957-4484/23/34/344003
18. Cook BS, Shamim A (2012) Inkjet printing of novel wideband and high gain antennas on low-cost paper substrate. IEEE Trans Antennas Propag 60:4148–4156. https://doi.org/10.1109/TAP.2012.2207079

19. Pang Y, Jian J, Tu T, Yang Z, Ling J, Li Y, Wang X, Qiao Y, Tian H, Yang Y, Ren T-L (2018) Wearable humidity sensor based on porous graphene network for respiration monitoring. Biosens Bioelectron 116:123–129. https://doi.org/10.1016/j.bios.2018.05.038
20. Wu J, Sun Y-M, Wu Z, Li X, Wang N, Tao K, Wang GP (2019) Carbon nanocoil-based fast-response and flexible humidity sensor for multifunctional applications. ACS Appl Mater Interfaces 11:4242–4251. https://doi.org/10.1021/acsami.8b18599
21. Zhu P, Liu Y, Fang Z, Kuang Y, Zhang Y, Peng C, Chen G (2019) Flexible and highly sensitive humidity sensor based on cellulose nanofibers and carbon nanotube composite film. Langmuir 35:4834–4842. https://doi.org/10.1021/acs.langmuir.8b04259
22. Dai H, Feng N, Li J, Zhang J, Li W (2019) Chemiresistive humidity sensor based on chitosan/zinc oxide/single-walled carbon nanotube composite film. Sens Actuators B Chem 283:786–792. https://doi.org/10.1016/j.snb.2018.12.056
23. Turkani VS, Maddipatla D, Narakathu BB, Saeed TS, Obare SO, Bazuin BJ, Atashbar MZ (2019) A highly sensitive printed humidity sensor based on a functionalized MWCNT/HEC composite for flexible electronics application. Nanoscale Adv 1:2311–2322. https://doi.org/10.1039/C9NA00179D

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