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Screen-printed graphene-carbon ink based disposable humidity sensor with wireless communication

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A B S T R A C T

Humidity sensing is crucial for several industrial, environmental, and healthcare applications, many of which require sensors in flexible form factors and with features such as disposability and facile fabrication processes. Herein, we present a flexible, cost-effective, and disposable humidity sensor developed on paper substrate. The screen-printed graphene-carbon (G-C) ink-based humidity sensor demonstrates good sensing performance in terms of change in resistance (~12.4 Ω/%RH) in humidity ranging from 25%RH to 91.7%RH. The sensor displays high flexibility (studied at bending radiuses 40 mm, 30 mm, 25 mm, and 20 mm), appreciable stability (> 4 months), high repeatability (> 100 cycles), short response/recovery time (~4 s/~6 s towards respiration rate monitoring) and good reproducibility (minor variations ~ ± 1 Ω/%RH). The efficacy of fabricated humidity sensor is evaluated for spatial humidity monitoring, respiration rate monitoring, and soil moisture monitoring. Finally, the real time monitoring of humidity is also demonstrated via wireless transmission of data to a smartphone to display the potential of the fabricated sensor for environmental, agricultural, and healthcare applications.

1. Introduction

Humidity plays a vital role in ecological, industrial, medical, and environmental monitoring (both indoor and outdoor) [1]. It is critical in sustaining biological life as deviations from comfortable humidity levels (humidity of ~30–60 % feels comfortable) could greatly impact the quality of life [2–4]. For example, humid air is harder to breathe and people suffering from asthma could feel short of breath or cough and wheeze [5]. The dust mites and moulds also tend to thrive at high humidity levels in the home [6]. Further, humidity also influences the agriculture farm productivity [4,7]. Therefore, the humidity sensors are needed in a wide range of indoor and outdoor applications ranging from smart buildings to agriculture, healthcare, and semiconductor and food packaging industries [8,9]. As a result, a wide variety of humidity sensors have been developed using capacitive, resistive, optical fibre, thermal, surface/bulk acoustic waves mechanisms etc. [1,10–13]. These sensors have been based on various nanomaterials, such as metal oxides [9,14], 2D nanomaterials and their composites such as SnO_2/RGO [15], graphene oxide/black phosphorus (GO/BP) [16], MoS_2/Ag [17], C_3N_4/Ag [18] nanoparticles etc. [19–21]. Whilst improvement in sensing characteristics (e.g., sensing range and sensitivity) have been achieved with a combination of these materials, their cumbersome synthesis and fabrication processes pose considerable challenge for their translation to flexible form factors. Emerging applications such as wearable systems, electronic require sensors on flexible substrates [22–27]. Further, simple and resource-efficient methods such as direct printing of sensitive materials on flexible substrates is desirable as these eventually helps to lower the cost of sensors and makes their disposability a viable option [28]. This is also needed in the light of growing health and environmental concerns related to electronic waste [29,30].

Herein, we present the miniaturised disposable humidity sensor developed on paper substrate (glossy photo paper, 150 gsm) by screen-printing the graphene-carbon ink. The developed humidity sensor is based on resistive sensing mechanism. Among different sensing technologies, resistive sensors have been considered advantageous considering their simple transduction mechanism, easy device integration and

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signal acquisition, easy manufacturing, cost-effective, small size, and long lifetime [31,32]. The biocompatible nature of the G-C ink with appreciable conductivity and a cheaper alternative to other metal-based ink formulations make it a viable choice for a disposable sensor. Amongst various available printing technologies, screen printing is closest to manufacturing and hence will make it easy to mass produce the sensors [33]. The ability to control the design, scale, and the notion of utilising various combinations of ink with varying functional material and substrates also make screen printing an attractive route. The choice of paper as a substrate for presented sensor is motivated by features such as recyclability, disposability nature, inexpensive, lightweight, and easy availability.

Although, paper itself can work as sensor for humidity monitoring [34–37], the slow response/recovery time and high baseline resistance (e.g., GΩ range) are the main downsides of using the paper as resistive humidity sensor. Slow response/recovery time could be suitable for applications such as environmental monitoring or packaging, where change in humidity is generally slow and gradual [36]. However, it is not desirable for application such as respiration rate monitoring, as targeted in the present work, where short response/recovery time is crucial to easily distinguish between breathing out/in cycles. Therefore, using paper as a substrate along with disposable, biocompatible and easily printable active humidity sensing materials could be the suitable choice for obtaining a disposable resistive humidity sensor, having desired sensing performance including short response and recovery times, appreciable sensitivity, good stability, repeatability, reproducibility etc. The humidity sensors developed in past with printed graphite ink on paper has limitation such as prone to cracking of electrodes [38]. Although such issues could be partly addressed by using conductive tapes (used as electrodes) [39], the integration issues could still make it challenging to push the work beyond lab. A wide variety of such sensors on paper, with different materials, fabrication techniques and sensing mechanisms have been reported in literature [40–43]. However, a flexible and disposable resistive humidity sensor targeting multifunction applications together demands further progress.

This work aims towards addressing these challenges through biocompatible graphene-carbon ink based screen-printed flexible and disposable resistive humidity sensor on paper substrate and their multifunctional applications (spatial humidity, respiration rate and soil humidity monitoring). Along with this, the impact of different substrates and number of printed layers on humidity sensing performance of the sensor is also analysed and discussed. The fabricated sensor displays good sensing performance in humidity range of 25%RH – 91.7%RH, excellent repeatability (> 100 cycles), stability (> 4 months) and short response/recovery time (∼4 s/∼6 s towards respiration rate monitoring). The biocompatible ink along with paper as the substrate makes an interesting combination for various environmental and healthcare applications. This has been demonstrated by using the fabricated sensor in real applications as well as real time humidity data transfer via wireless communication system. Further, to evaluate the usage of presented sensor in real applications, the performance is also evaluated under different bending (at bending radiuses 40, 30, 25 and 20 mm) and twisting (at angles 45° and 90°) conditions. The obtained results indicates that the presented humidity sensor is robust enough for wearable and flexible electronic applications.

2. Materials and methods

2.1. Materials

Graphene-carbon ink (C2171023D1:Graphene Carbon Ink:BG04, Sun Chemical), substrates (glossy paper, matt paper and sylvicta), and dielectric ink (D2070423P5:Grey Dielectric Ink:BG04, Sun Chemical) were used.

2.2. Sensor fabrication using screen printing and characterisation

The graphene-carbon based humidity sensors were fabricated on glossy paper (glossy photo paper, 150 gsm), matt paper (matt double sided photopaper, 240 gsm) and sylvicta (220 gsm) substrates using the screen-printing technique (*Screen Stencil Printer C920 from AUREL Automation*). The number of screen-printed layers on glossy paper i.e*.,* single layer (sensor 1), double layer (sensor 2), and triple layer (sensor 3) were optimised based on the humidity sensing performance of the sensor 1, sensor 2, and sensor 3. For double and triple layer configurations, each printed layer was dried before printing next layer. Further, heat treatment was given to the as printed sensors for 1 hr at 60 °C. The connections for 2-wire resistance monitoring were made and wiring followed by dielectric placement on contact points. The characterisation details are provided in Supporting Note 1. Further, the humidity sensing performance was analysed as discussed in later section.

2.3. Humidity sensing set-up

The humidity sensing characteristic analysis of the sensor was performed by placing the sensor inside inhouse build sensing chamber. The complete details about sensing chamber are provided in Supporting Note 2. The change in resistance behaviour of the sensor under different humidity levels was measured using the digital multimeter (Agilent 34461 A 6½ Digit Multimeter) and recorded using the LabVIEW interface. The dehumidification process was obtained via purging the sensing chamber and exposing the sensor to air. The sensing set-up is shown in Fig. S1. The schematic illustration of the of the fabrication process and humidity sensing setup is depicted in Fig. 1(a-b).

3. Results and discussion

3.1. Material characterisation

The screen-printed graphene-carbon sensing layer-based humidity sensor is depicted in Fig. 2(a). The surface morphology of the sensing layer is observed using scanning electron microscopy (SEM). Fig. 2(b-c) shows SEM images at two different scale bars i.e*.,* 5 µm and 500 nm. The carbon particles appear to embellish the graphene sheets and randomly distributed G-C network of the sensing layer is observed. The SEM images of the sensing layer at 100 µm and 50 µm are shown in the Fig. S2. The diffractogram of the printed layer is displayed in Fig. 2(d). The peak observed at ∼26.3° corresponds to the (002) plane, typically observed for carbon [44]. The hydrophilicity analysis is paramount for humidity sensors as hydrophilic nature of the sensing layer has significant impact on the sensing properties [45]. Excessive or deficient hydrophilicity is not desired for humidity sensors as it may lead to slow recovery or response of the sensor. Therefore, moderate hydrophilicity of the sensing layer could be advantageous to obtain rapid response/recovery. Herein, contact angle measurement is performed using sessile drop method to analyse the hydrophilicity of the printed G-C sensing layer as displayed in Fig. 2(e). The droplet on the sensing layer surface formed a contact angle of 66.6°. A material is considered hydrophilic if the contact angle is < 90° i.e., lower the angle, higher the hydrophilicity [39, 46,47]. Hence, the observed contact angle shows the moderate hydrophilicity of the sensing layer. Further, the thickness of the single, double, and triple G-C layer is observed using cross-sectional SEM image analysis as shown in Fig. S3. The layer thickness is found to be ∼10 µm, ∼16 µm and ∼22 µm, for single, double, and triple layer, respectively.

3.2. Influence of substrate on humidity sensing performance

The choice of substrate is important for development of humidity sensor specifically when paper is used as a substrate. The substrate features such as surface roughness, porosity, contact angle etc. play a significant role in defining the properties of screen-printed layer as well

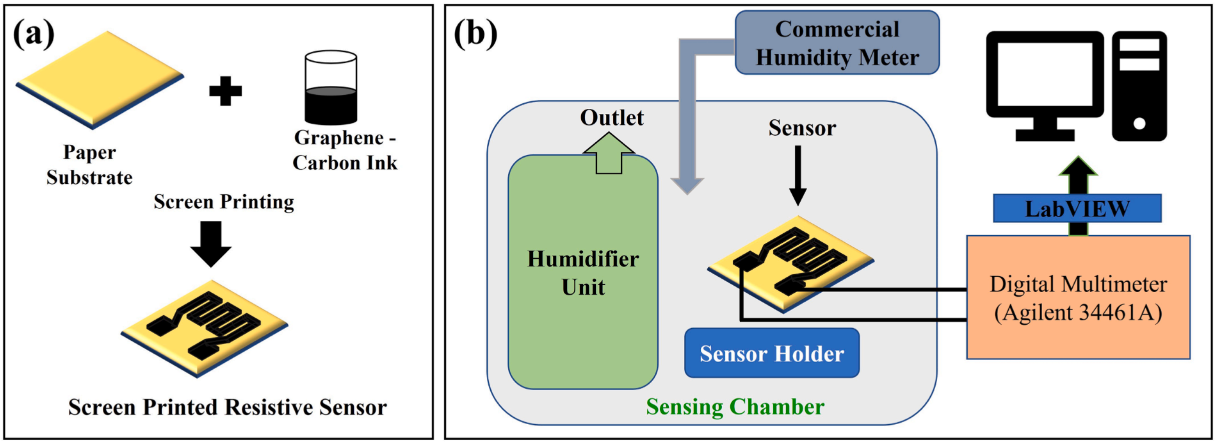

Fig. 1. Schematic illustration of the (a) sensor fabrication by screen printing of graphene-carbon ink on paper substrate and (b) sensing setup for humidity sensing measurements.

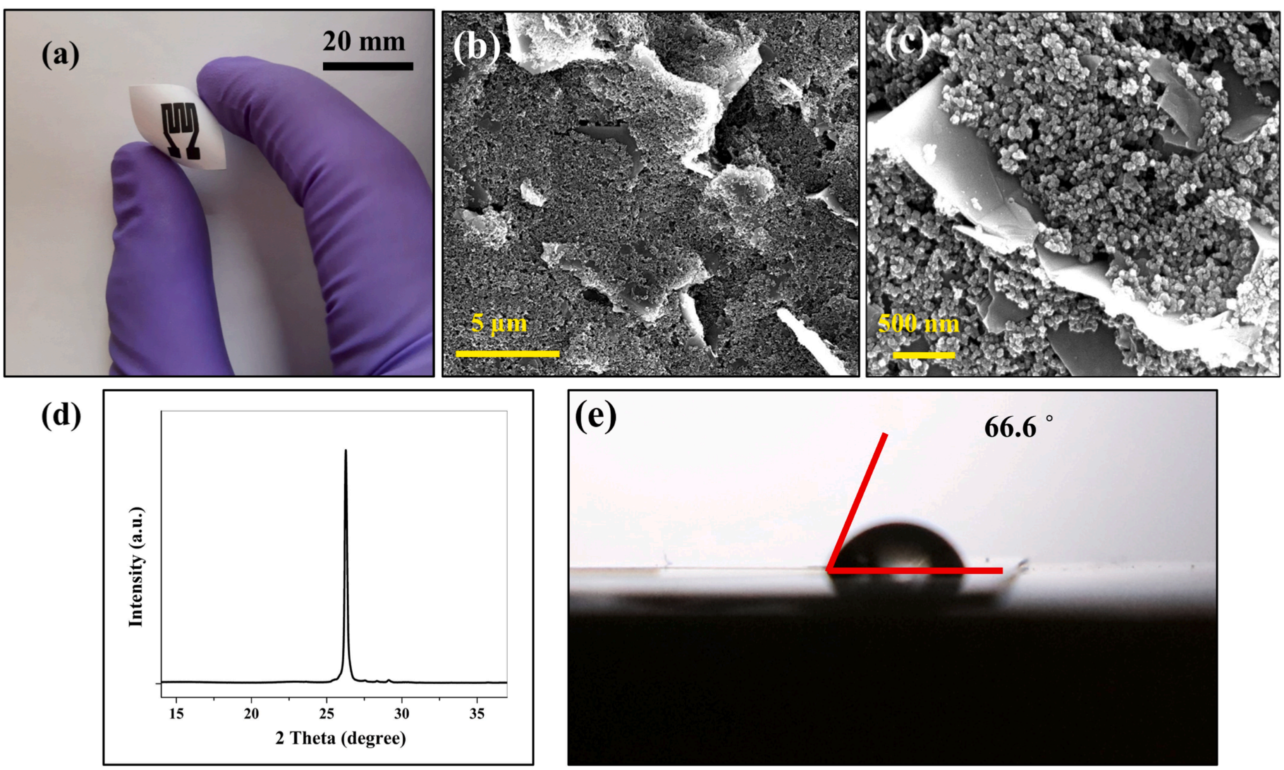

Fig. 2. (a) Screen-printed G-C sensing layer-based humidity sensor. SEM images of the G-C sensing layer at (b) 5 µm and (c) 500 nm scales; (d) XRD of the G-C sensing layer and (e) Contact angle measurement of the sensing layer.

the sensing characteristics of the sensor [42]. To understand the impact of paper substrate properties on printing and humidity sensing performance, we have considered three different types of paper substrates i.e., glossy photopaper, matt photopaper and sylvicta (which is a translucent, functional barrier paper). The detailed discussion on influence of substrate on humidity sensing performance (refer Fig. S4, Fig. S5, Fig. S6 and Fig. S7) is explained in Supporting Note 3. Based on the obtained results, it is observed that the paper substrate properties not only influenced the printed layer quality (shown in Fig. S4) but also affected the humidity sensing performance (shown in Fig. S7) of the developed sensors as discussed in Supporting Note 3.

Further, the obtained results suggests that sensors printed on matt paper and sylvicta substrates could be used for applications (e.g., environmental monitoring or packaging) where humidity changes comparatively slowly. However, considering respiration rate monitoring as one of the targeted applications of the present work, the humidity sensors having short response/recovery time is vital to distinguish between breath out/in cycles. Therefore, sensor printed on glossy paper substrate is selected for further experiments considering its shorter response/recovery time as compared to sensors printed on matt paper and sylvicta substrates. Moreover, along with the substrate study, the impact of number of screen-printed G-C layers on the humidity sensing performance of the selected sensor is also analysed and discussed in the next section.

3.3. *Humidity sensing performance*

To understand the impact of number of printed layers on humidity sensing, the performance of sensors printed on glossy paper substrates (sensor 1 i.e., single G-C layer, sensor 2 i.e., double G-C layer, and sensor 3 i.e., triple G-C layer) is analysed in humidity range ~ 35%RH to ~ 91% RH. The baseline resistance is found to be different in sensor 1, 2 and 3, which is ascribed to single, double, and triple screen-printed G-C layer configuration. The change in the resistance of all three sensors under humid conditions is shown in Fig. 3(a). It is observed that resistance of sensors increases with increase in percentage of relative humidity (% RH). The mechanism for this change in resistance with humidity is discussed in later section. The performance of the sensors is compared by measuring the change in resistance (ΔR) with change in %RH (Δ% RH) as shown in Fig. 3(b). The ΔR/Δ%RH is observed as 12.88 Ω/%RH, 10.36 Ω/%RH and 2.32 Ω/%RH, for sensor 1, sensor 2 and sensor 3, respectively, for humidity range of ~ 35%RH to ~ 91%RH. Observed results indicate superior performance for single G-C layer printed sensor i.e., sensor 1 as compared to sensor 2 and sensor 3, in terms of change in resistance for the measured humidity range. Also, response/recovery time of the sensors are analysed and compared as shown in Fig. 3(c). The response and recovery times are determined as time taken by the sensor to attain ~ 90% of maximum resistance change under exposure to humidity and dehumidification, respectively. The response/recovery times are observed as ~31 s/~8 s, ~90 s/~10 s and ~150 s/~20 s for sensor 1, sensor 2 and sensor 3, respectively. The shortest response and recovery times is observed for sensor 1 as compared to sensor 2 and sensor 3. Thus, making it more suitable for humidity sensing applications. This comparatively short response/recovery time of sensor 1 may be explained based on the sensing layer configuration as single layer sensor (sensor 1) exhibits higher specific surface area or surface to volume ratio as compared to sensor 2 and sensor 3. Therefore, on comparing the humidity sensing performances of all the three sensors, sensor 1 based on single G-C layer is found as the optimised humidity sensor. Further characterisations and application studies are measured using sensor 1 and hence would be referred as humidity sensor, unless otherwise stated.

The humidity sensing characteristics of sensor 1 are analysed in the range of 25%RH to 91.7%RH at room temperature (25 ± 2 ℃). The resistance of sensing layer at 25%RH is considered as baseline resistance (R_B) of the sensor. The resistance vs time graph, as shown in Fig. 4(a), represents the change in resistance behaviour of the sensor at different humidity levels in the range of ~25%RH to ~91.7%RH. Further, the % response (% R) of the humidity sensor is determined at each intermediate humidity level. The % R value is calculated as follows:

$$\frac{\Delta R}{R_B} \times 100 \qquad (1)$$

Where, $\Delta R = R_H - R_B$; R_H is resistance at specific humidity level; R_B is baseline resistance of the sensor analysed at 25%RH. The % response vs % RH graph is shown in Fig. 4(b). The lowest observed % response value is 1.31% at 30%RH, which gradually increases up to 43.16 % at 91.7% RH. The change in response seems to stagnate after ~89%RH and does not show significant alterations even at higher humidity levels. Therefore, the sensor response could be found more suitable until the range of ~90%RH. The repeatability characteristic of the humidity sensor is shown in Fig. 4(c). The repeatability performance is analysed within a humidity range of 35%RH to 91%RH. The five cyclic measurements are highly repeatable in nature with nearly similar response and recovery

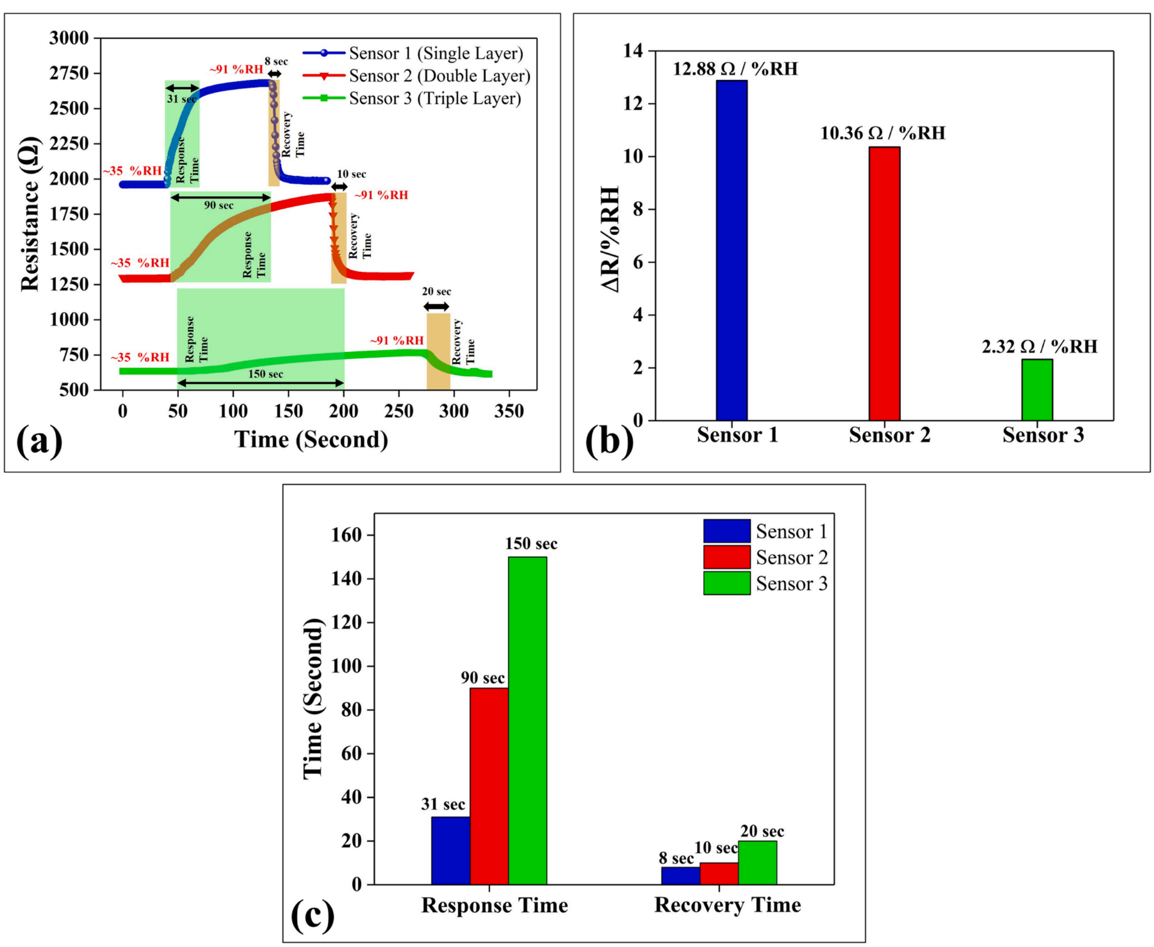

Fig. 3. Comparative analysis of humidity sensing properties of sensor 1, sensor 2 and sensor 3. (a) Dynamic response characteristics of sensor 1, 2 and 3 for humidity range 35%RH to 91%RH. (b) Change in resistance per %RH analysis. (c) Response and recovery times analysis.

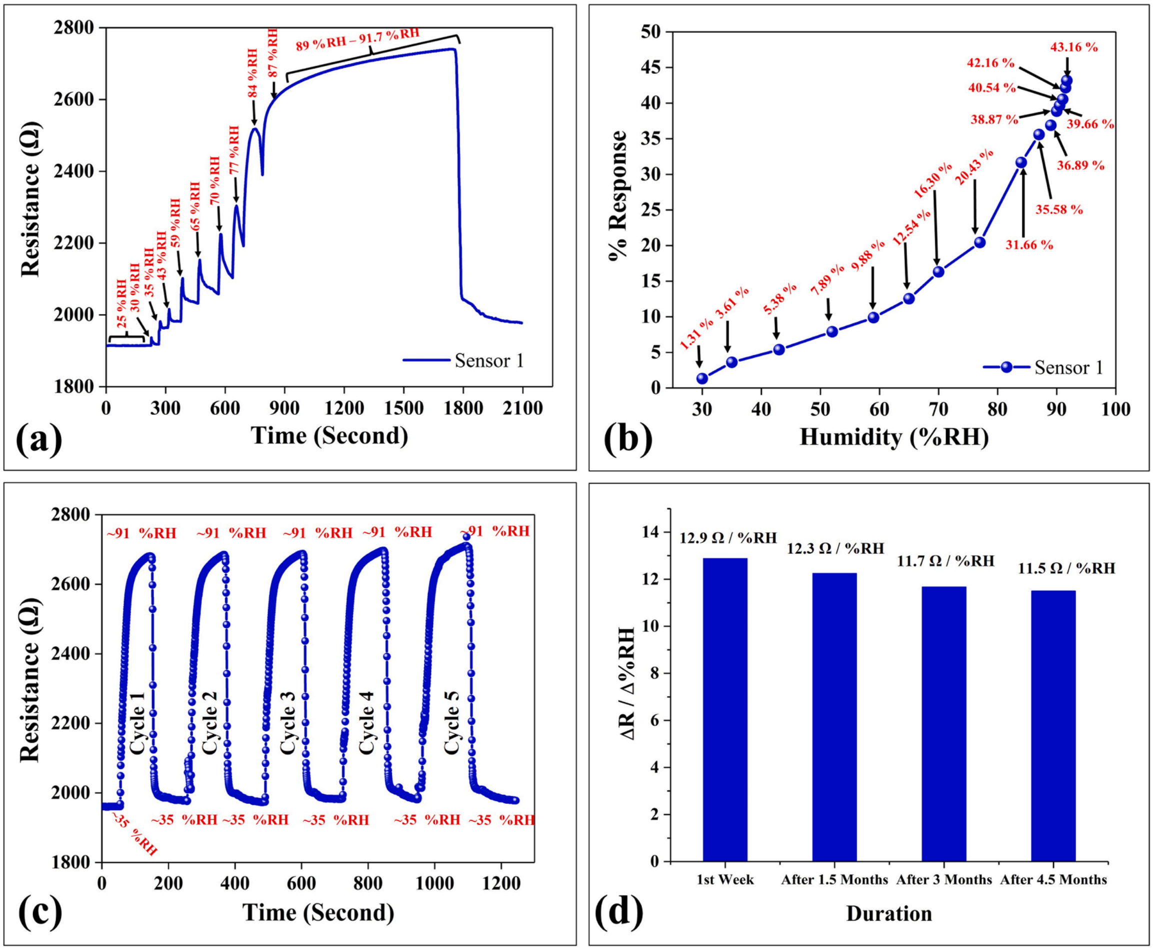

Fig. 4. Humidity sensing performance of sensor 1 (a) Dynamic response characteristics i.e., resistance vs time graph for humidity range within 25%RH to 91.7%RH. (b) % Response analysis for humidity range within 25%RH to 91.7%RH and (c) Cyclic repeatability analysis. (d) Stability analysis by analysing the sensor response from 1st week to 4.5 months.

times (∼31–35 s/∼7–8 s) as explained previously. The stability of the fabricated sensor is examined for more than 4 months by monitoring its performance at regular intervals. The response of the sensor is observed in first week (12.88 Ω/%RH) and compared with the response obtained after 1.5 months (12.25 Ω/%RH), 3 months (11.67 Ω/%RH) and 4.5 months (11.50 Ω/%RH) as shown in the Fig. 4(d). The comparative analysis shows that the performance of the sensor is reasonably stable even after 4.5 months with a minor drop (∼1.4 Ω/%RH) in sensing performance. Further, the reproducibility of the sensor is also analysed by repeating the fabrication route. The response of the reproduced sensors (RS1, RS2 and RS3) is compared with the original sensor as shown in the Fig. S8. The response of the reproduced sensors is found well in accordance with the original sensor and show only minor variations (∼ ± 1 Ω/%RH). This displays the potential applicability of the developed sensor for multiple measurements.

3.4. Sensor applications

3.4.1. Respiration rate monitoring

Respiration rate monitoring is a low-cost, non-invasive, and useful approach that can be used for real-time physiological monitoring and early illness diagnosis [48,49]. Considering the humidity sensing performance of the fabricated sensor, potential application towards human respiration or breathing rate monitoring is investigated [50]. The presence of humidity in the exhaled human breath can be used for monitoring the breathing rate per minute. It is important to monitor the breathing rate specially for the patients suffering from severe illness or diseases such as chronic obstructive pulmonary disease (COPD), asthma, bronchiectasis etc. [49,51]. The normal breathing rate for a healthy adult at rest is 12–20 breaths per minutes (bpm) [51]. Whereas breathing rate > 24 bpm for several hours is and indicator of health risk [52]. Breathing rate > 27 bpm is considered as an important sign for the cardiac arrest in hospitals [53]. Moreover, continuous breathing rate monitoring during sleep is important to identify the sleep apnoea condition [38]. Hence, breathing rate monitoring with presented sensor can help in various healthcare assessments.

The fabricated humidity sensor is found suitable for continuous breathing monitoring. The relative humidity in exhaled breath generally lies in/around the range of ∼40–90 % [54]. Although, nasal breathing is usual way of breathing for human, under certain circumstances (e.g., strenuous exercises, nasal congestion due to cold etc.) mouth breathing is also utilised [55]. Therefore, both nose as well as mouth breathing are considered for breathing rate monitoring in the present work. As shown in Fig. 5(a), the humidity sensor is used for three breaths out and in (exhale/inhale) cyclic measurements towards mouth breathing. Humidity in exhaled mouth breath is found to be ∼ 83%RH (monitored using commercial humidity meter). The calculated humidity value (monitored using fabricated sensor) based on change in resistance comes out to be ∼ 80%RH (i.e., accuracy ± 3%RH) with short response/recovery time (∼4 s/∼6 s). Supplementary video S1 provides an understanding about sensor response towards five breaths out/in cyclic measurements. Further, the mouth breathing is continuously monitored for 5 min for an adult at rest condition, as shown in Fig. 5(b). The sensor response towards continuous breathing monitoring is found similar to

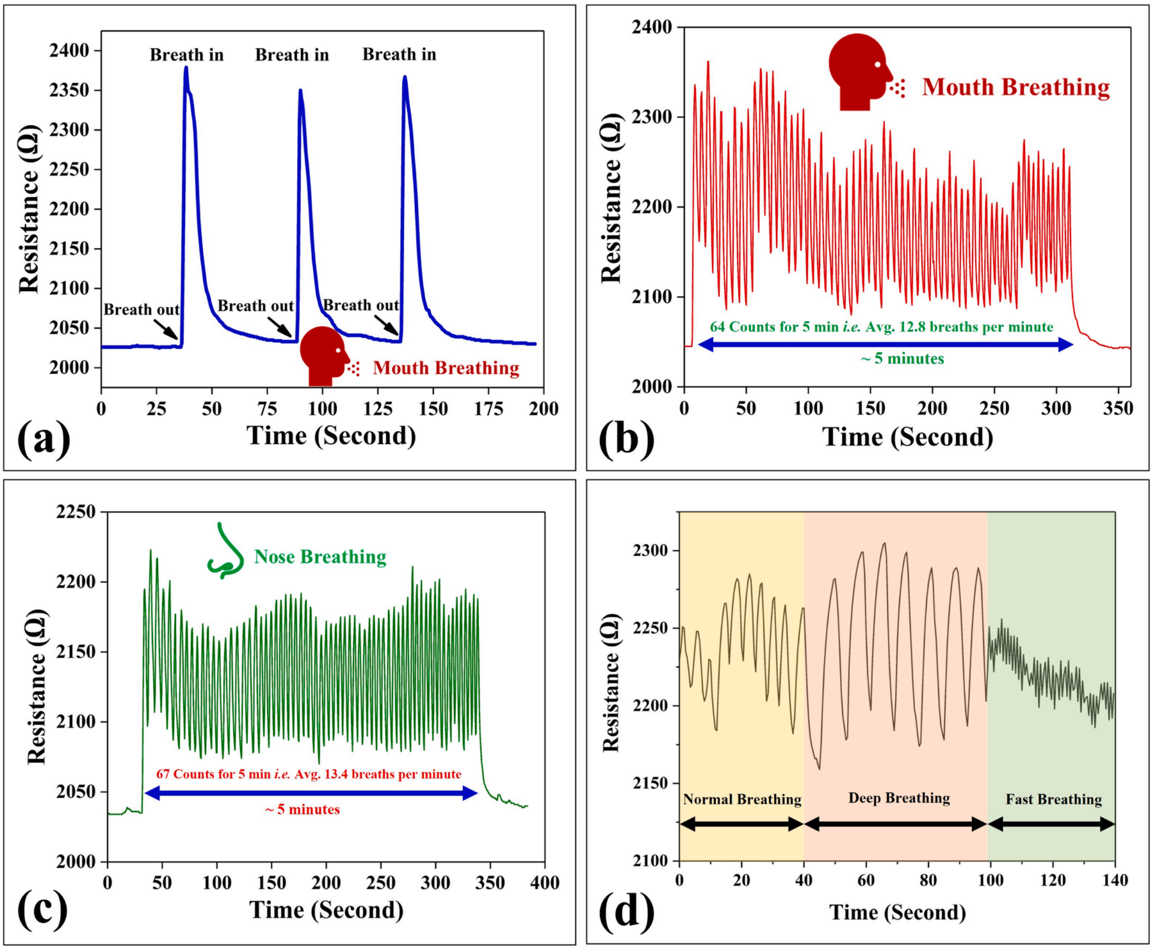

Fig. 5. Humidity sensor application (a) Three breaths out/in cyclic measurements for mouth breath humidity monitoring. (b) Continuous mouth breathing monitoring for 5 min. (c) Continuous nose breathing monitoring for 5 min. (d) Sensor's response towards the normal, deep, and fast breathing patterns.

the breaths out/in cyclic measurements and peak corresponding to each breath out/in cycle can be easily distinguished. However, the sensor is not able to return to its baseline resistance value as recovery time of the sensor is greater than the interval between the breaths. Furthermore, the nose breathing is also continuously monitored for another 5 min as shown in the Fig. 5(c). Exhaled nose breath humidity is found to be ~ 72%RH (monitored using commercial humidity meter). The lower %RH level in exhaled nasal breath as compared to mouth breath could be ascribed to absence of saliva in nasal breathing. The calculated humidity value (monitored using fabricated sensor) based on change in resistance comes out to be ~ 70%RH with short response/recovery time. The sensor is showing distinguishable response towards continuous breathing monitoring (mouth as well as nose breathing) corresponding to each exhaled breath out/in cycle. This displays the applicability of the fabricated sensor towards breathing rate monitoring.

Supplementary material related to this article can be found online at doi:10.1016/j.snb.2022.132731.

Further, the response of the sensor is also examined for different breathing patterns such as normal, deep, and fast breathing. A comparative analysis of the normal, deep, and fast breathing patterns is depicted in Fig. 5(d). The observed results indicate the ability of the sensor to clearly distinguish among the observed responses towards different patterns. Moreover, the cyclic repeatability of the sensor is also analysed by continuously monitoring the response for > 100 breathing cycles as shown in Fig. S9. The obtained response clearly shows highly repeatable behaviour of the sensor towards exhaled breath. Hence, the obtained results display the applicability of the fabricated sensor towards breathing rate monitoring and open avenues for its utilisation in health monitoring applications.

3.4.2. *Soil moisture monitoring*

Soil moisture or humidity level is an important parameter for the development and growth of plants/crops. Limited or excessive soil moisture can lead to plant death or root diseases [56]. Optimum moisture level plays a significant role for adequate plant growth and enhanced crop productivity. Therefore, regular monitoring of soil moisture/humidity level is necessary. The fabricated sensor is tested for the compost soil moisture level monitoring as shown in Fig. 6. Initially, the sensor is placed under normal ambient conditions and resistance of the sensor is monitored. Then, the sensor is placed on the surface of the compost soil sample and change in resistance is monitored. Supplementary video S2 displays the sensor's response towards soil moisture monitoring. The detailed discussion on soil moisture monitoring is presented in Supporting Note 4. The obtained results indicate the suitability of the developed resistive humidity sensor towards soil moisture monitoring applications.

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3.5. *Bending and twisting study*

The flexibility or bending ability is another key feature needed to evaluate the robustness and hence the suitability of the sensor in wearable systems, some of which are used in applications discussed above. Therefore, bending, and twisting tests are carried out on fabricated humidity sensor as displayed in the Fig. 7. The performance of the

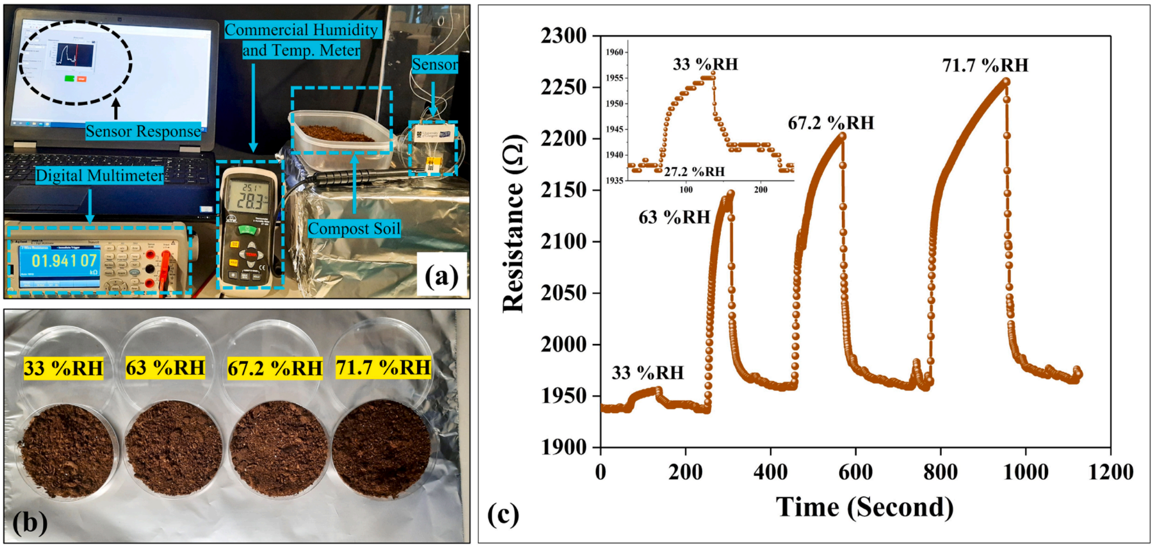

Fig. 6. Humidity sensor application towards compost soil moisture analysis (a) Set-up for monitoring the soil moisture level. (b) Prepared soil samples with different moisture levels. (c) Sensing characteristics of the sensor towards prepared samples.

sensor is examined under different bending conditions. The sensor is bent at a radius of 40 mm, 30 mm, 25 mm, and 20 mm as shown in Fig. 7 (a-d). A slight variation (increase in resistance, < 6 %) in baseline resistance is observed for the considered bending radiuses which may be due to change in conductivity under expansion of G-C layer. To understand the impact of bending conditions on humidity sensing performance, the sensor is exposed to similar humidity level (3 breaths cyclic measurements) before and under bending condition at considered bending radius as shown in the Fig. 7(e-h). The obtained response (in terms of change in resistance) in both the cases i.e., prior to bending and under bending, is observed to be comparable for all the considered bending radii. Further, the sensor is exposed to 100 bending cycles at 40 mm radius as shown in the Fig. 7(i). The humidity sensing performance of the sensor is examined before and after 100 bending cycles and found to be nearly similar. Supplementary video S3 displays the bending test of the humidity sensor at 40 mm bending radius. Similarly, twisting study is also carried out for the fabricated sensor. Fig. 7(j-l) depicts the sensor under normal and twisted (at angle 45° and 90°) conditions. A slight variation (decrease in resistance, < 3 %) in baseline resistance is observed under the considered twisted conditions which may be due to change in conductivity due to compression of G-C layer. The sensor is exposed to similar humidity level (single breath cycle measurement) under normal and twisted conditions and the response is analysed as shown in Fig. 7(m). The obtained response (in terms of change in resistance) in all the cases i.e., prior to twisting and under twisting (at angle 45° and 90°), is found to be comparable. Therefore, the obtained results (under different bending and twisting conditions) show that presented humidity sensor is robust enough for wearable and flexible electronic applications.

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3.6. Real time sensing and monitoring of humidity level

The IoT-enabled electronic systems are rapidly appearing in healthcare, consumer products, security, and agriculture sectors. Various protocols (e.g., Zigbee, Wi-Fi, Bluetooth, LoRa etc.) are used in these applications to enable real-time wireless data communication between transmitters and receivers [57,58]. Herein, the wireless humidity level monitoring system is developed using the Node MCU ESP8266 microcontroller enabled with the Wi-Fi protocol. As shown in Fig. 8, the wireless system consists of a fabricated humidity sensor, a data processing unit, an inbuilt Wi-Fi module for data transmission, and a power supply unit. The electronic system is operated through a ~3 V supply, and a 32-bit microcontroller incorporated with an ESP-12 module (Wi-Fi SoC). Arduino IDE platform is used to write the software module of the humidity monitoring system. The control code is built using embedded C programming language. Further, the source code is generated and uploaded to the microcontroller, the monitoring system is able to detect the real-time humidity levels with a small delay of around 15 s. ThingSpeak (IoT analytics platform) is used to aggregate and visualise the live data streams in the cloud display. Real time humidity monitoring system is depicted in the Fig. 8(a). The detected humidity data is received and displayed in real time on a smartphone. Fig. 8(b) represents the real time monitored humidity level i.e., humidity vs time graph in the front-end visualisation, which clearly represents both previously recorded and the real time humidity measurements. While Fig. 8(c) represents the geographical information of the measurement spot of the humidity sensor along with an easy display highlighting the humidity level. Supplementary video S4 displays the real time sensing using human breathing monitoring, where the humidity at RT is measured as 30%RH in the application reading, which increases to 82%RH under exposure to exhaled mouth breath.

Supplementary material related to this article can be found online at doi:10.1016/j.snb.2022.132731.

3.7. Sensing mechanism and disposability analysis

The resistance of the sensor increases with the increase in the humidity level. The change in these values across the given range could be ascribed to the formation of a physisorbed layer of water molecules on the surface of the sensing material at lower %RH values. Fig. 9 displays the plausible mechanism to explain the change in resistance observed across different humidity value. Moreover, the detailed discussion on sensing mechanism and disposability analysis is presented in Supporting Note 5. Further, Table S1 provides a comparative analysis of the present work with state of the art. The table compare various features such as material used, fabrication technique applied, flexibility, disposability, and the humidity range measured. The state-of-the-art discussion suggest the combined features of disposability and flexibility for an excellent resistive humidity sensor suitable for multifunctional applications as reported in this work.

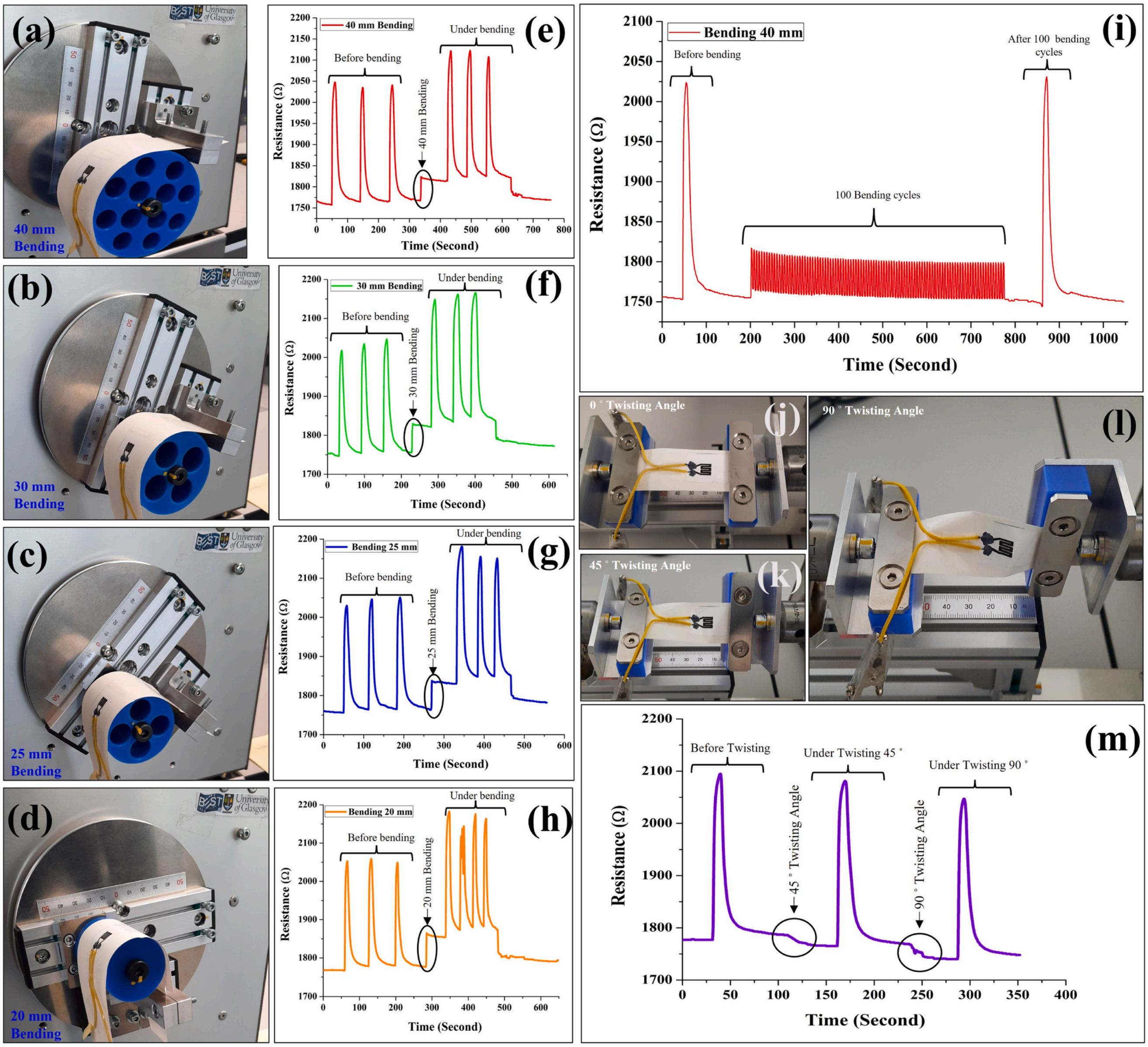

Fig. 7. Bending and twisting studies of the humidity sensor. Sensor under (a) 40 mm (b) 30 mm (c) 25 mm and (d) 20 mm bending conditions. Response analysis (3 cyclic measurements) prior to and under (e) 40 mm (f) 30 mm (g) 25 mm and (h) 20 mm bending conditions. (i) Sensor performance before and after 100 bending cycles. Sensor under (j) normal condition i.e., 0° twisted (k) 45° twisted and (l) 90° twisted condition. (m) Sensor response under normal, 45° and 90° twisting conditions.

4. Conclusion and future scope

The flexible and disposable humidity sensor presented in this paper exhibits good sensing performance (~12.4 Ω/%RH) in range 25%RH to 91.7%RH. The observed characteristics of the sensor such as high flexibility (studied at multiple bending radii), high repeatability (>100 cycles), appreciable stability (>4 months), short response/recovery time (~4 s/~6 s towards respiration rate monitoring) and good reproducibility (with minor variations ~ ± 1 Ω/%RH) makes it suitable for multiple applications. The wireless transmission of data could further open new avenues towards remote monitoring applications. Beyond measuring humidity, the sensor could also be explored to monitor the strain and temperature change caused in the environment and could be coupled as a multifunctional sensory patch [59]. The use of paper as a substrate in the current work restricts its application to rather dry environment. However, similar electrode pattern printed on disposable polymeric sheets can evade those issues. The present wireless transmission system powered by a ~3 V supply. This drives up the cost of the disposable sensor and restricts the overall integration process. Thus, integrating sensors with more energy efficient technologies can solve the present issue. Energy harvesters along with energy storage devices can potentially pave the route to such independent sensory modules.

CRediT authorship contribution statement

Ajay Beniwal: Conceptualization, Methodology, Investigation, Formal analysis, Validation, Writing – original draft. **Priyanka Ganguly:** Methodology, Investigation, Formal analysis. **Akshaya Kumar Aliyana:** Software, Investigation. **Gaurav Khandelwal:** Investigation. **Ravinder Dahiya:** Conceptualization, Writing – review & editing, Formal analysis, Funding acquisition, Supervision. All the authors contributed towards manuscript writing.

Declaration of Competing Interest

The authors declare that they have no known competing financial

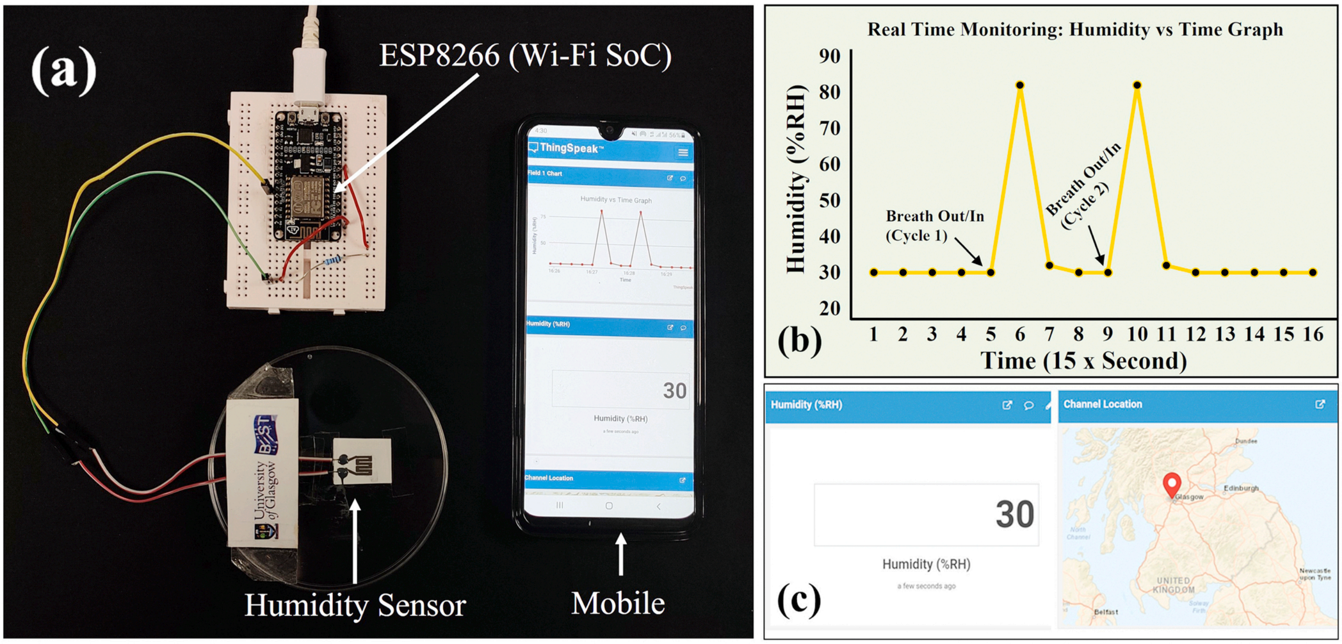

Fig. 8. (a) Real time humidity sensing system; (b) the displayed data showa the change in the humidity values at multiple cycles, and (c) Data displaying the geographical location of the measurement spot.

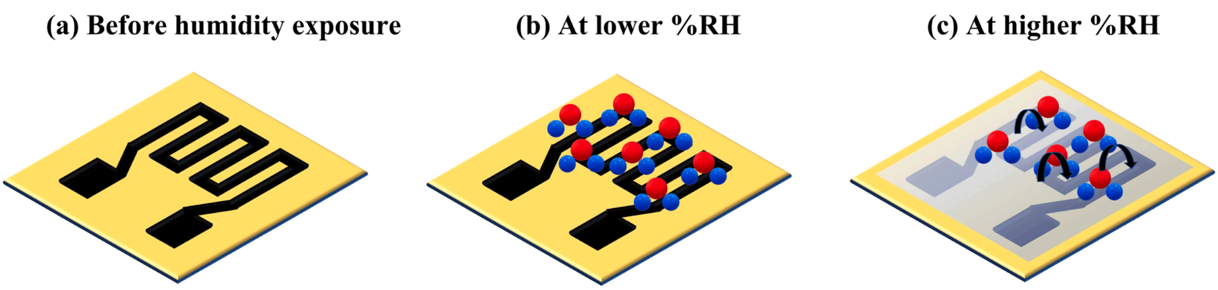

Fig. 9. Schematic illustration of the sensing mechanism representing humidity sensor (a) before humidity exposure, (b) at lower %RH and (c) higher %RH exposure.

interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

Data will be made available on request.

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Supporting information

Supporting information contains Supporting Notes presenting detailed discussion about the characterisation, sensing set-up, influence of the different substrates on humidity sensing characteristics, sensor application towards soil humidity monitoring, sensing mechanism and disposability analysis. Supporting information contains experimental set-up for humidity sensing analysis and SEM images of the G-C sensing layer at 100 µm and 50 µm scales. Cross-sectional SEM images for layer thickness analysis is presented. Contact angle vs time graph for glossy, matt and sylvicta substrates is given. Cyclic repeatability (> 100 cycles) and reproducibility analysis graphs are also presented. Sensor degradation study throughout the 8 weeks is displayed. Supplementary videos are presented displaying breaths out/in cyclic measurements, soil moisture/humidity monitoring, bending test of the humidity sensor, and real time humidity sensing. Further, comparative analysis of the present work with state of the art is also presented.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.snb.2022.132731.

References

[1] R. Malik, V.K. Tomer, V. Chaudhary, M.S. Dahiya, A. Sharma, S. Nehra, et al., An excellent humidity sensor based on In–SnO_2 loaded mesoporous graphitic carbon nitride, J. Mater. Chem. A 5 (2017) 14134–14143.
[2] Y. Wang, L. Zhang, Z. Zhang, P. Sun, H. Chen, High-sensitivity wearable and flexible humidity sensor based on graphene oxide/non-woven fabric for respiration monitoring, Langmuir 36 (2020) 9443–9448.
[3] K. Takei, W. Gao, C. Wang, A. Javey, Physical and chemical sensing with electronic skin, Proc. IEEE 107 (2019) 2155–2167.

[4] L. Manjakkal, F.F. Franco, A. Pullanchiyodan, M. González-Jiménez, R. Dahiya, Natural jute fibre-based supercapacitors and sensors for eco-friendly energy autonomous systems, Adv. Sustain. Syst. 5 (2021), 2000286.
[5] R. Beasley, A. Semprini, E.A. Mitchell, Risk factors for asthma: is prevention possible? Lancet 386 (2015) 1075–1085.
[6] L.G. Arlian, J.S. Neal, M.S. Morgan, D.L. Vyszenski-Moher, C.M. Rapp, A. K. Alexander, Reducing relative humidity is a practical way to control dust mites and their allergens in homes in temperate climates, J. Allergy Clin. Immunol. 107 (2001) 99–104.
[7] F. Romero, S. Cazzato, F. Walder, S. Vogelgsang, S.F. Bender, M.G. van der Heijden, Humidity and high temperature are important for predicting fungal disease outbreaks worldwide, New Phytol. 234 (2022) 1553–1556.
[8] Z. Duan, Y. Jiang, H. Tai, Recent advances in humidity sensor for human body related humidity detections, J. Mater. Chem. C 9 (2021) 14963–14980.
[9] M. Parthibavarman, V. Hariharan, C. Sekar, High-sensitivity humidity sensor based on SnO_2 nanoparticles synthesized by microwave irradiation method, Mater. Sci. Eng. C 31 (2011) 840–844.
[10] J. Li, T. Fu, Y. Chen, B. Guan, M. Zhuo, T. Yang, et al., Highly sensitive humidity sensors based on Sb-doped $ZnSnO_3$ nanoparticles with very small sizes, Cryst. Eng. Commun. 16 (2014) 2977–2983.
[11] X. Yao, Y. Cui, A. PEDOT, PSS functionalized capacitive sensor for humidity, Measurement 160 (2020), 107782.
[12] M. Velumani, S. Meher, Z. Alex, Impedometric humidity sensing characteristics of SnO_2 thin films and SnO_2–ZnO composite thin films grown by magnetron sputtering, J. Mater. Sci. Mater. Electron. 29 (2018) 3999–4010.
[13] W. Liu, H. Qu, J. Hu, W. Pang, H. Zhang, X. Duan, A highly sensitive humidity sensor based on ultrahigh-frequency microelectromechanical resonator coated with nano-assembled polyelectrolyte thin films, Micromachines 8 (2017) 116.
[14] S. Nagaraju, A.S. Roy, J. Kumar, K.R. Anilkumar, G. Ramagopal, Humidity sensing properties of surface modified polyaniline metal oxide composites, J. Eng. 2014 (2014), 925020.
[15] D. Zhang, H. Chang, P. Li, R. Liu, Q. Xue, Fabrication and characterization of an ultrasensitive humidity sensor based on metal oxide/graphene hybrid nanocomposite, Sens. Actuators B Chem. 225 (2016) 233–240.
[16] P. He, J. Brent, H. Ding, J. Yang, D. Lewis, P. O'Brien, et al., Fully printed high performance humidity sensors based on two-dimensional materials, Nanoscale 10 (2018) 5599–5606.
[17] N. Li, X.-D. Chen, X.-P. Chen, X. Ding, X. Zhao, Ultra-high sensitivity humidity sensor based on MoS_2/Ag composite films, IEEE Electron Device Lett. 38 (2017) 806–809.
[18] V.K. Tomer, N. Thangaraj, S. Gahlot, K. Kailasam, Cubic mesoporous Ag@ CN: a high performance humidity sensor, Nanoscale 8 (2016) 19794–19803.
[19] M. Soni, M. Bhattacharjee, M. Ntagios, R. Dahiya, Printed temperature sensor based on PEDOT: PSS-graphene oxide composite, IEEE Sens. J. 20 (2020) 7525–7531.
[20] S. Dervin, P. Ganguly, R. Dahiya, Disposable electrochemical sensor using Graphene oxide–chitosan modified carbon-based electrodes for the detection of tyrosine, IEEE Sens. J. 21 (2021) 26226–26233.
[21] F. Liu, S. Deswal, A. Christou, Y. Sandamirskaya, M. Kaboli, R. Dahiya, Neuro-inspired electronic skin for robots, Sci. Robot. 7 (2022), eabl7344.
[22] M. Shojaei Baghini, A. Vilouras, M. Douthwaite, P. Georgiou, R. Dahiya, Ultra-thin ISFET-based sensing systems, Electrochem. Sci. Adv. (2021), e2100202.
[23] O. Ozioko R. Dahiya. Smart tactile gloves for haptic interaction, communication, and rehabilitation, advanced intelligent systems. 2021 2100091 doi: 10.1002/elsa.202100202.
[24] R. Dahiya, E-skin: from humanoids to humans [point of view], Proc. IEEE 107 (2019) 247–252.
[25] Y. Pang, J. Jian, T. Tu, Z. Yang, J. Ling, Y. Li, et al., Wearable humidity sensor based on porous graphene network for respiration monitoring, Biosens. Bioelectron. 116 (2018) 123–129.
[26] O. Ozioko, P. Karipoth, M. Hersh, R. Dahiya, Wearable assistive tactile communication interface based on integrated touch sensors and actuators, IEEE Trans. Neural Syst. Rehabil. Eng. 28 (2020) 1344–1352.
[27] Y. Su, W. Li, L. Yuan, C. Chen, H. Pan, G. Xie, et al., Piezoelectric fiber composites with polydopamine interfacial layer for self-powered wearable biomonitoring, Nano Energy 89 (2021), 106321.
[28] A. Benchirouf, S. Palaniyappan, R. Ramalingame, P. Raghunandan, T. Jagemann, C. Müller, et al., Electrical properties of multi-walled carbon nanotubes/PEDOT: PSS nanocomposites thin films under temperature and humidity effects, Sens. Actuators B Chem. 224 (2016) 344–350.
[29] E.S. Hosseini, S. Dervin, P. Ganguly, R. Dahiya, Biodegradable materials for sustainable health monitoring devices, ACS Appl. Bio Mater. 4 (2020) 163–194.
[30] M. Chakraborty, J. Kettle, R. Dahiya, Electronic waste reduction through devices and printed circuit boards designed for circularity, IEEE Journal on Flexible, IEEE J. Flex. Electron. 1 (2022) 4–23.
[31] E. Assunção da Silva, C. Duc, N. Redon, J.-L. Wojkiewicz, Humidity sensor based on PEO/PEDOT:PSS blends for breath monitoring, Macromol. Mater. Eng. 306 (2021), 2100489.
[32] C. Chen, M. Jiang, X. Luo, H. Tai, Y. Jiang, M. Yang, et al., Ni-Co-P hollow nanobricks enabled humidity sensor for respiratory analysis and human-machine interfacing, Sens. Actuators B Chem. 370 (2022), 132441.
[33] Y. Kumaresan, O. Ozioko, R. Dahiya, Multifunctional electronic skin with a stack of temperature and pressure sensor arrays, IEEE Sens. J. 21 (2021) 26243–26251.
[34] D. Barmpakos, A. Segkos, C. Tsamis, G. Kaltsas, A disposable flexible humidity sensor directly printed on paper for medical applications, J. Phys. Conf. Ser. 931 (2017), 012003.
[35] G. Niarchos, G. Dubourg, G. Afroudakis, M. Georgopoulos, V. Tsouti, E. Makarona, et al., Humidity sensing properties of paper substrates and their passivation with ZnO nanoparticles for sensor applications, Sensors 17 (2017) 516.
[36] C. Gaspar, J. Olkkonen, S. Passoja, M. Smolander, Paper as active layer in inkjet-printed capacitive humidity sensors, Sensors 17 (2017) 1464.
[37] Y. Feng, L. Xie, Q. Chen, L. Zheng, Low-cost printed chipless RFID humidity sensor tag for intelligent packaging, IEEE Sens. J. 15 (2015) 3201–3208.
[38] F. Güder, A. Ainla, J. Redston, B. Mosadegh, A. Glavan, T. Martin, et al., Paper-based electrical respiration sensor, Angew. Chem. Int. Ed. 55 (2016) 5727–5732.
[39] Z. Duan, Y. Jiang, M. Yan, S. Wang, Z. Yuan, Q. Zhao, et al., Facile, flexible, cost-saving, and environment-friendly paper-based humidity sensor for multifunctional applications, ACS Appl. Mater. Interfaces 11 (2019) 21840–21849.
[40] R. Alrammouz, J. Podlecki, A. Vena, R. Garcia, P. Abboud, R. Habchi, et al., Highly porous and flexible capacitive humidity sensor based on self-assembled graphene oxide sheets on a paper substrate, Sens. Actuators B Chem. 298 (2019), 126892.
[41] J.-W. Han, B. Kim, J. Li, M. Meyyappan, Carbon nanotube based humidity sensor on cellulose paper, J. Phys. Chem. C 116 (2012) 22094–22097.
[42] X. Zhang, D. He, Q. Yang, M.Z. Atashbar, Rapid, highly sensitive, and highly repeatable printed porous paper humidity sensor, Chem. Eng. J. 433 (2022), 133751.
[43] R.M. Morais, Md.S. Klem, G.L. Nogueira, T.C. Gomes, N. Alves, Low cost humidity sensor based on PANI/PEDOT:PSS printed on paper, IEEE Sens. J. 18 (2018) 2647–2651.
[44] F. Hof, K. Kampioti, K. Huang, C. Jaillet, A. Derré, P. Poulin, et al., Conductive inks of graphitic nanoparticles from a sustainable carbon feedstock, Carbon 111 (2017) 142–149.
[45] Z. Wei, J. Huang, W. Chen, Q. Huang, Fabrication and characterization of flexible capacitive humidity sensors based on graphene oxide on porous PTFE substrates, Sensors 21 (2021) 5118.
[46] X. Feng, L. Feng, M. Jin, J. Zhai, L. Jiang, D. Zhu, Reversible super-hydrophobicity to super-hydrophilicity transition of aligned ZnO nanorod films, J. Am. Chem. Soc. 126 (2004) 62–63.
[47] X. Yu, Z. Wang, Y. Jiang, F. Shi, X. Zhang, Reversible pH-responsive surface: From superhydrophobicity to superhydrophilicity, Adv. Mater. 17 (2005) 1289–1293.
[48] Y. Su, G. Chen, C. Chen, Q. Gong, G. Xie, M. Yao, et al., Self-powered respiration monitoring enabled by a triboelectric nanogenerator, Adv. Mater. 33 (2021), 2101262.
[49] B. Liu, A. Libanori, Y. Zhou, X. Xiao, G. Xie, X. Zhao, et al., Simultaneous biomechanical and biochemical monitoring for self-powered breath analysis, ACS Appl. Mater. Interfaces 14 (2022) 7301–7310.
[50] B. Li, G. Xiao, F. Liu, Y. Qiao, C.M. Li, Z. Lu, A flexible humidity sensor based on silk fabrics for human respiration monitoring, J. Mater. Chem. C 6 (2018) 4549–4554.
[51] B. Hill, S.H. Annesley, Monitoring respiratory rate in adults, Br. J. Nurs. 29 (2020) 12–16.
[52] M.A. Cretikos, R. Bellomo, K. Hillman, J. Chen, S. Finfer, A. Flabouris, Respiratory rate: the neglected vital sign, Med. J. Aust. 188 (2008) 657–659.
[53] J.F. Fieselmann, M.S. Hendryx, C.M. Helms, D.S. Wakefield, Respiratory rate predicts cardiopulmonary arrest for internal medicine inpatients, J. Gen. Intern. Med. 8 (1993) 354–360.
[54] E. Mansour, R. Vishinkin, S. Rihet, W. Saliba, F. Fish, P. Sarfati, et al., Measurement of temperature and relative humidity in exhaled breath, Sens. Actuators B: Chem. 304 (2020), 127371.
[55] J. Zhang, X.-X. Wang, B. Zhang, S. Ramakrishna, M. Yu, J.-W. Ma, et al., In situ assembly of well-dispersed Ag nanoparticles throughout electrospun alginate nanofibers for monitoring human breath – smart fabrics, ACS Appl. Mater. Interfaces 10 (2018) 19863–19870.
[56] E.A.A.D. Nagahage, I.S.P. Nagahage, T. Fujino, Calibration and validation of a low-cost capacitive moisture sensor to integrate the automated soil moisture monitoring system, Agriculture 9 (2019) 141.
[57] M.J. Christoe, J. Han, K. Kalantar-Zadeh, Telecommunications and data processing in flexible electronic systems, Adv. Mater. Technol. 5 (2020), 1900733.
[58] M.J. Christoe, J. Yuan, A. Michael, K. Kalantar-Zadeh, Bluetooth signal attenuation analysis in human body tissue analogues, IEEE Access 9 (2021) 85144–85150.
[59] Y. Lu, K. Xu, L. Zhang, M. Deguchi, H. Shishido, T. Arie, et al., Multimodal plant healthcare flexible sensor system, ACS Nano 14 (2020) 10966–10975.

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