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Research article

Ultrasensitive stretchable bimodal sensor based on novel elastomer and ionic liquid for temperature and humidity detection

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A B S T R A C T

In this work, we present a novel stretchable bimodal sensor that can simultaneously detect temperature and humidity changes based on polyhydroxyethyl acrylate (PHEA) elastomer infused with 1-ethyl-3-methylimidazolium tetrafluoroborate (EMIM-BF4) ionic liquid. The sensor exhibits high transparency, stability, and biocompatibility, as well as excellent mechanical and sensing properties. The sensor can achieve a maximum strain of 761%, a sensitivity of 4.5%/°C at room temperature, a detection range from −35 to 120 °C, and a response time of 10 ms. The sensor is able to provide acute response to movement of human hand at close range and can detect temperature changes as small as 0.004 °C in the range of 20–30 °C. The sensor also responds to humidity change, showing a high sensitivity to humidity change of 4.4%/RH% under the temperature of 30 °C. The sensor can be used for various applications in wearable electronics, human-machine interfaces, and soft robotics.

1. Introduction

Stretchable sensors that can detect multiple stimuli such as temperature and humidity are highly desirable for applications in wearable electronics, human-machine interfaces, and soft robotics. Temperature sensing is especially important for monitoring human health and physiological activities, as well as environmental changes [1–4]. However, most of the existing stretchable sensors are based on electronic conductors and semiconductors, which have limitations in transparency, stretchability, stability, and biocompatibility [5–8].

Ionic conductors, on the other hand, offer advantages such as high transparency, low impedance, and good biocompatibility [9–11]. However, they often face challenges due to the high ion concentration and the presence of water in the matrix for ion dissolution, which can result in poor mechanical properties and water evaporation [12–14]. Therefore, there is a pressing need to develop novel ionic conductors that can combine high stretchability, transparency, stability, and sensitivity for bimodal sensing.

Various attempts have been made to create sensors based on hydrogels and conducting elements, utilizing different measurement techniques [6,15,16]. However, many of these sensors rely on complex structures or incorporate conducting particles, such as multiwall carbon nanofibers, rendering the material opaque and compromising its mechanical properties [17]. Additionally, some previous works involved the use of metallic salts, introducing the challenge of corrosion over extended periods, which can damage the polymer structure [18,19]. In 2018, S. Ding and colleagues [20] introduced a non-corroding, single-phased ion gel material that has high sensitivity to temperature. Nonetheless, their utilization of a hydrophobic matrix limited the material's capability to simultaneously detect moisture. There also have been other trials to make stretchable temperature and humidity sensors by application of stretchable designs [21,22] , but by incorporating such designs, the manufacturing process becomes more complicated. Consequently, the development of a bimodal sensor capable of sensing both temperature and humidity remain an unsolved challenge.

The elastomer PHEA (Poly Hydroxyl Acrylate) catches our attention due to its superior hydrophilicity and biocompatibility. The

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hydrophilicity of PHEA allows it to absorb water molecule from ambient air and to develop a humidity sensing mechanism based on it. And the high biocompatibility of PHEA allows long period of contact of PHEA matrix to human skin, which allows the fabrication of breath sensors from it. In addition, the high biodegradability of PHEA will cause little environmental impact upon commercialization.

1-Ethyl-3-Methylimidazolium Tetrafluoroborate (EMI-BF4), as a novel kind of ionic liquid, has been used in lithium batteries to improve their ionic conductivity and ion transportation behaviors at room/low temperatures [23]. However, the utilization of EMI-BF4 as an ionic conductor when coupled with hydrogel remains a relatively unexplored area of research.

In this work, we report an ultrasensitive stretchable bimodal sensor that can sense temperature and humidity simultaneously based on novel elastomer and ionic liquid samples. The elastomer can provide mechanical strength and elasticity to the sensor. During mechanical property tests, PHEA (Poly Hydroxyl Acrylate) sample doped with 10 wt% ionic liquid is capable of a maximum strain of 761%. The ionic liquid infusion provides exquisite sensing ability to both temperature and humidity change, with a high sensitivity of 4.5%/°C at room temperature range. wide detection ranges from −35 to 120 °C, fast response time of 10 ms. These characteristics render it well-suited for a variety of applications in soft electronics.

2. Materials and methods

2.1. Material and equipment

Material: Monomer: Hydroxyl Acrylate (HEA), ionic liquid: 1-Ethyl-3-Methylimidazolium Tetrafluoroborate (EMI-BF4), light absorber: Diphenyl(2,4,6-trimethylbenzoyl) phosphine oxide (TPO) are all brought from Sigma Aldrich.

Equipment: Fluke 2638A Source meter, Wan Hao pulling machine, Mark 10 F305-IM Universal Testing machine. EZ4 Laboratory compact spin coater, Seville LED UV Black Light.

2.2. Preparation of sensor piece

The sample was created through a series of steps. Initially, a resin mixture was prepared by blending a hydroxyl acrylate monomer with EMI-BF4 ionic liquid (in varying concentrations from 5 to 40 wt%) along with 0.5 wt% TPO photo initiator. This mixture was then applied to a mica substrate using spin coating. As shown in Fig. 1a, the subsequent polymerization of the resin film was achieved under UV light, and the sensor piece was carefully removed, followed by a thorough ethanol wash. The resulting sensor samples consistently had a thickness of 0.1 mm. A post-curing process was carried out under UV light to enhance the sensor's mechanical properties.

2.3. Characterization of the sensor piece

The temperature and humidity of the samples were characterized through a series of tests. For temperature response analysis, a PHEA sample underwent baking at 80 °C until it reached a 10 wt% weight loss. Subsequently, the strip was sealed with PTFE tape, and its resistance changes with increasing temperature were recorded. Cooling tests involved lowering the strip to subzero temperatures using liquid nitrogen, recording resistance changes during the cooling process. These datasets were processed to construct a comprehensive temperature-response curve.

Humidity response measurements consisted of sealing a bare sensing strip within a container containing a water source and hygroscopic salt (Lithium Chloride). Resistance changes were recorded as humidity gradually decreased. This process was repeated at

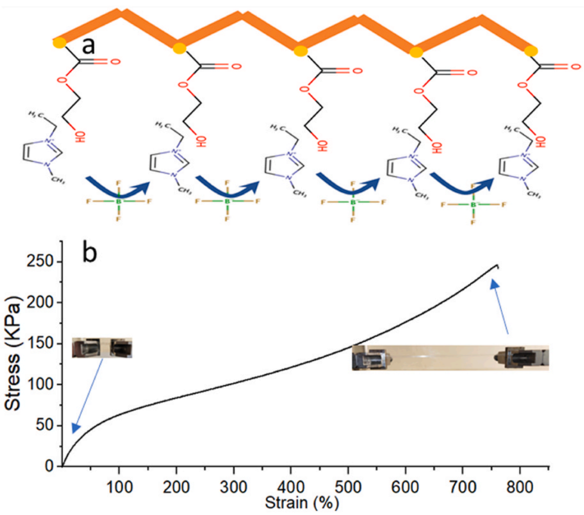

Fig. 1. (a)Max tensile strength of the polymer. (b) Structural schematic of the polymer.

various temperatures (30, 40, 50, 60, and 80 °C), generating Resistance-Humidity matrices for different temperature levels.

Mechanical property assessments were conducted using a pulling test, and the resulting data were processed using specialized software. For breath sensing, a PHEA sample sealed with PTFE tape was attached to an N95 mask's surface to monitor temperature changes resulting from human breath.

To determine the gauge factor, a pulling machine is used to gradually apply strain to standard rectangular-shaped PHEA samples, with resistance changes recorded. In motion sensing, the same piece of PHEA used for breath sensing was affixed to a lab door and integrated into the sensing system. The door's magnet was part of the system to automatically release when a sharp resistance drop was detected.

2.4. Data collection

Resistance values were read using a source meter and converted into ambient temperature data for temperature measurements. For humidity measurements, the first strip's ambient temperature reading was used to determine humidity levels by incorporating the resistance of the second strip into a Temperature-Humidity (T-H) matrix.

3. Results and discussion

3.1. Mechanical properties of the polymer

The initially fabricated PHEA (Poly Hydroxyl Acrylate) samples demonstrated commendable mechanical properties, with remarkable stretchability exceeding 790%. This underscores PHEA's suitability as a high-quality elastomer. In our work, we incorporated various ionic liquids, including EMI-BF4 (1-Ethyl-3-Methylimidazolium Tetrafluoroborate), to impart conductivity. Notably, the addition of a relatively modest amount of ionic liquid within the range of 10–20 wt% had a minimal impact on mechanical properties. As depicted in Fig. 1b, the elastomer enriched with 10 wt% EMI-BF4 exhibited an outstanding maximum stretchability of 761%.

3.2. General properties of the material

We doped the PHEA samples with various ionic liquids, each at 10 wt%, and compared their respective room temperature resistivity values. We used an undoped PHEA sample as a reference, which exhibited insulating properties with a resistivity of 1.5×10^7 Ω/m. After doping with different ionic liquids, the resistivity of the samples decreased, with some falling into the resistivity range of semiconductors. As depicted in Fig. 2a, the resistivity of the samples doped with 10 wt% EMI-BF4 was compared with those doped with EMI-TFSI (1-Ethyl-3-methylimidazolium bis (trifluoromethyl sulfonyl) imide) and EMIES (1-Ethyl-3-methylimidazolium ethyl sulfate), which are commonly used ionic liquids. The results revealed that the addition of EMI-BF4 led to the lowest resistivity among the three samples at room temperature (25 °C) under an ambient relative humidity of RH = 80%. Notably, increasing the EMI-BF4 content to 20–30 wt% had minimal impact on mechanical properties, ensuring high elasticity was maintained.

As reported in previous studies [24,25], conducting elastomers can serve as effective strain sensors. We selected the PHEA samples doped with the 10 wt% EMI-BF4 to do the test. Subsequently, we measured the gauge factor of this sample, as depicted in Fig. 2b, the figure illustrates that the resistivity of the sample varies in response to elongation (strain) up to 200% with a gauge factor of 5.7.

3.3. Electric properties of the material

Our initial research objective was to measure resistivity concerning temperature for temperature sensing applications. As shown in Fig. 3a, our findings revealed that the selected sample (PHEA doped with 10 wt% EMI-BF4) maintained conductivity at low temperatures down to −35 °C and remained stable even at elevated temperatures up to 120 °C. Notably, the sample retained relatively low

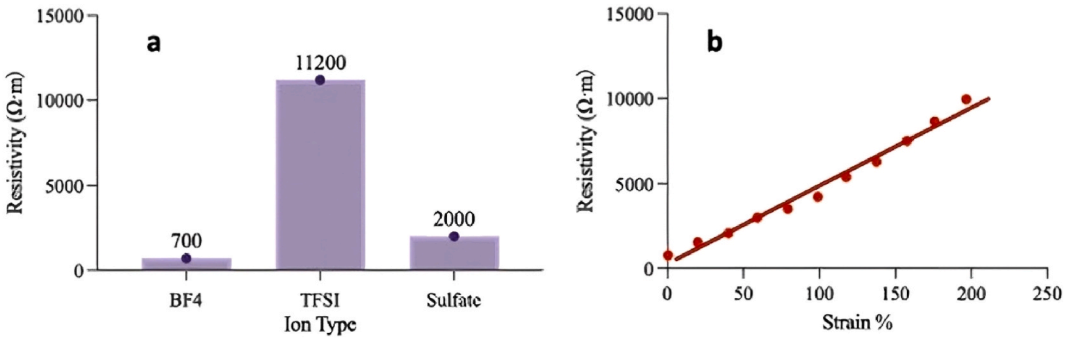

Fig. 2. (a)The resistivity of HEA ionogels with different ionic liquid. (b)Strain-resistivity curve of the material.

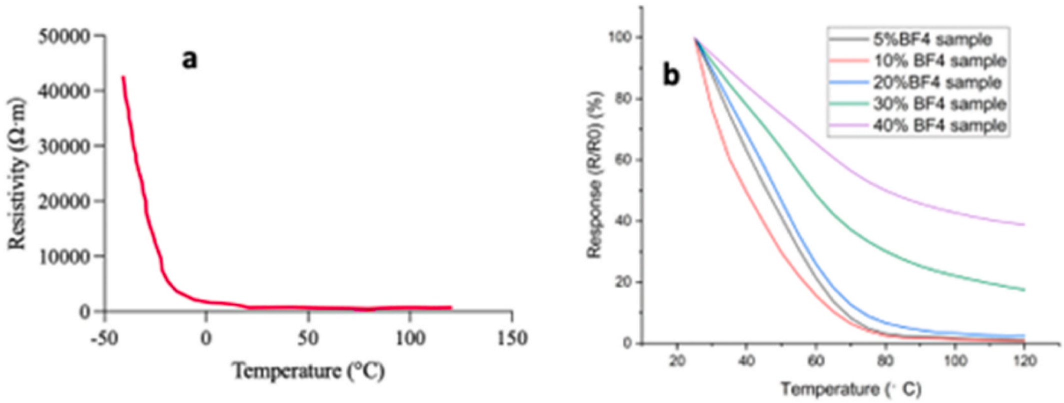

Fig. 3. (a)The upper and lower limit temperature sensing range of EIL. (b)The response to temperature of sample with different EMI-BF4 concentration.

resistivity even at a low temperature of −20 °C. This temperature response closely aligns with similar behavior observed in other previously reported conducting hydrogels [26,27]. Furthermore, when compared to existing materials of the same class [6,18], our sample exhibited a broader sensing range.

In addition to its wide temperature stability range, the sample also exhibited high sensitivity to temperature changes. Fig. 3b illustrates the varying responses of differently EMI-BF4 concentration samples (ranging from 5% to 40%) to a temperature change from 25 to 120 °C. To maintain consistent water content during these tests, a hydrophobic tape (Polytetrafluoroethylene) was used to seal the samples after pretreatment. It is evident that the sample with 10 wt% EMI-BF4 displayed the highest sensitivity to temperature changes, while the 40 wt% sample exhibited the least sensitivity. To quantitatively measure the sensitivity, we defined the thermal response as the relative resistance variation (ΔR/R0) concerning the initial resistance level (R0). Sensitivity was calculated from the slope of the fitted response-versus-temperature curve using Equation 1.

$$S(\%) = (\Delta R)/(R0 * \Delta T) * 100\% \quad \text{Eq.1}$$

The sample with 10 wt% EMI-BF4 showcased the highest sensitivity at 4.5%/°C at room temperature (25 °C), surpassing most previously reported materials [28–37,46]. A detailed comparison is provided in Table S1. However, it is important to note that the sample with 5 wt% EMI-BF4 exhibited lower sensitivity (2.5%) compared to the 10 wt% sample (4.5%). This observation can be attributed to the nature of PHEA as a long-chain elastomer, which undergoes coiling and relaxation as temperature changes. When the dopant concentration is reduced to very low levels, the polymer network formed by these coiled chains becomes denser, potentially inducing cross-links [31]. This significantly reduces the ability of conducting ions to move within the polymer, thus reducing sensitivity. Conversely, with very high dopant concentrations, the restriction on ion movement inside the polymer network is minimal at room temperature, resulting in little change as temperature varies.

After studying the samples' response to the temperature change, we then conducted research on its repeatability of results. First the PHEA sample doped with 10 wt% EMI-BF4 is deprived of water content and sealed with PTFE tape as mentioned before and situated in an air tight box. Then the temperature inside the box is heated from 70 to 100 °C. As shown in Fig. 4, the sample is able to reproduce the same resistance value at a given temperature after a few cycles. This points out that the sample has reproductible resistivity values

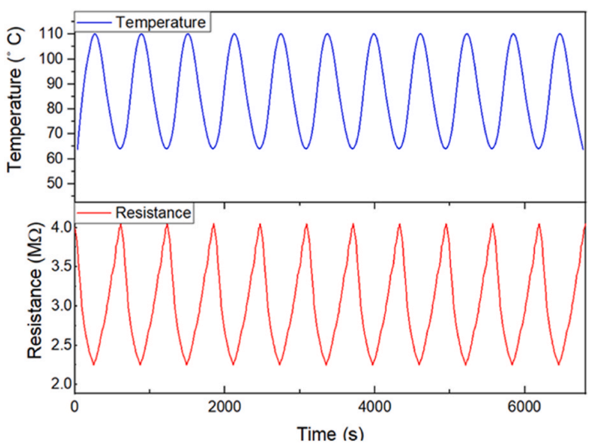

Fig. 4. Cyclic temperature and resistance behavior of EIL.

when subjected to a slow cyclic temperature change.

Our next study was to test the response of the sample to changes in humidity at a specific temperature. As illustrated in Fig. 5a, the PHEA sample doped with 10 wt% EMI-BF4 attains various equilibrium weights when exposed to different relative humidity levels, ranging from 20% to 80%, at room temperature. Subsequently, we measured the resistivity of the sample at room temperature as the ambient humidity increased from 30% to 80%, with results depicted in Fig. 5b. As shown in the figure, there is a substantial change in resistivity as the environmental humidity increases from 30% to 80%.

The sample is then subjected to humidity change under different temperatures from 30 to 80 °C as shown in Fig. 6. The data collected is arrayed into a temperature humidity matrix comprised of resistance curves under different temperature. The matrix is later used in the fabrication of the bimodal sensor. From the TH-matrix data, it's found that the sample has a highest sensitivity of 4.4%/RH % under the temperature of 30 °C, which is 3 times more than previously reported resistive moisture sensors, which have their sensitivities in the range of 1–2%/RH% at maximum [38–41]. Detailed comparison is shown in Table S2.

3.4. Potential applications of the material

Based on the properties of the PHEA sample doped with 10 wt% EMI-BF4, we have developed a bimodal sensor capable of simultaneously sensing temperature and humidity. By detecting the movement of a human hand and monitoring the behavior of a lit candle, our sensor has demonstrated high sensitivity to both temperature and humidity. After this, we have extended the applications to motion detection and respiratory sensing. From the tests, it is shown that the sample has the potential to be used in various areas.

3.5. Bimodal sensor

As stated in the "Introduction" section, the bimodal sensor is made of one sealed and one bare PHEA strip doped with 10 wt% EMI-BF4. The strips are situated on a transparent plastic substrate. The circuit is then connected to the substrate. In a typical measuring cycle, the resistance value of the sealed strip is measured first. Then the resistance value is substituted into the data curve shown in Fig. 3a to acquire the temperature value. After acquiring the temperature value, the resistance value of the bare strip is measured. Then the resistance value of the bare strip is substituted into the data curves shown in Fig. 6 together with the temperature value acquired above to calculate the humidity. Then the combined temperature and humidity data is sent to the screen by a Wi-Fi module. A photo and schematic of the bimodal sensor is shown in Fig. 7.

Our sensor boasts an exceptional temperature resolution, capable of detecting temperature changes as small as 0.004 °C, at the temperature range from 20 to 30 °C. This stands in stark contrast to the majority of commercial products (Table S3) and academically reported stretchable sensors, which typically exhibit resolutions within the range of 0.1–0.5 °C [42,43].

We first demonstrated this high resolution by employing the temperature sensor to detect a human hand. We placed the hand at a distance of 15 cm from the sensor and detecting the temperature is 27.648 °C (see movie S1). As the hand gradually approached to a distance of 5 cm, the temperature changed to 27.664 °C. From the test data depicted in Fig. 8, it is evident that the bimodal sensor consistently generated temperature change signals throughout this process and could clearly discern the movement of the hand, thus affirming the high temperature resolution. A comparison of cost of our sensor with commercial sensors that have a similar resolution is shown in Table S4. It is shown that, the elastomer sensor costs much less to produce than existing high-resolution sensors.

In addition to its remarkable temperature resolution, the bimodal sensor also exhibits a high humidity resolution. We conducted a candle test to compare our bimodal sensor with the commercial temperature and humidity dual sensor, DHT22 (see movie S2). As shown in Fig. 9, we firstly placed a lit candle 3 cm in front of the EIL sensor, with the L value set to zero. Then, we gradually moved the candle to L = 15 cm and recorded the changes in sensor output during this process. It is noticeable that the DHT22 failed to detect humidity changes during this phase, maintaining a stable humidity output of 81.6%. However, our bimodal sensor demonstrated humidity variations ranging from 81.592% to 81.605%, showing our robust humidity sensing capabilities. Simultaneously, in comparison to the DHT22, our bimodal sensor was able to precisely detect detailed temperature changes during this test.

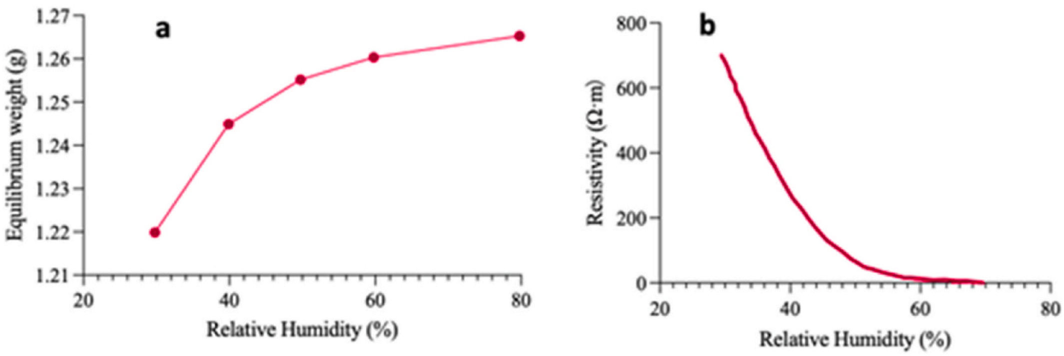

Fig. 5. (a)The equilibrium weight with the increase of humidity at room temperature. (b)The resistivity versus humidity curve of the sensor at room temperature.

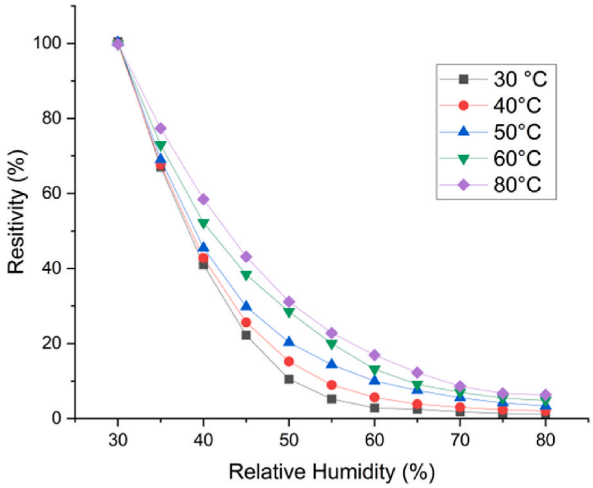

Fig. 6. The response to humidity of the sensor at different temperature.

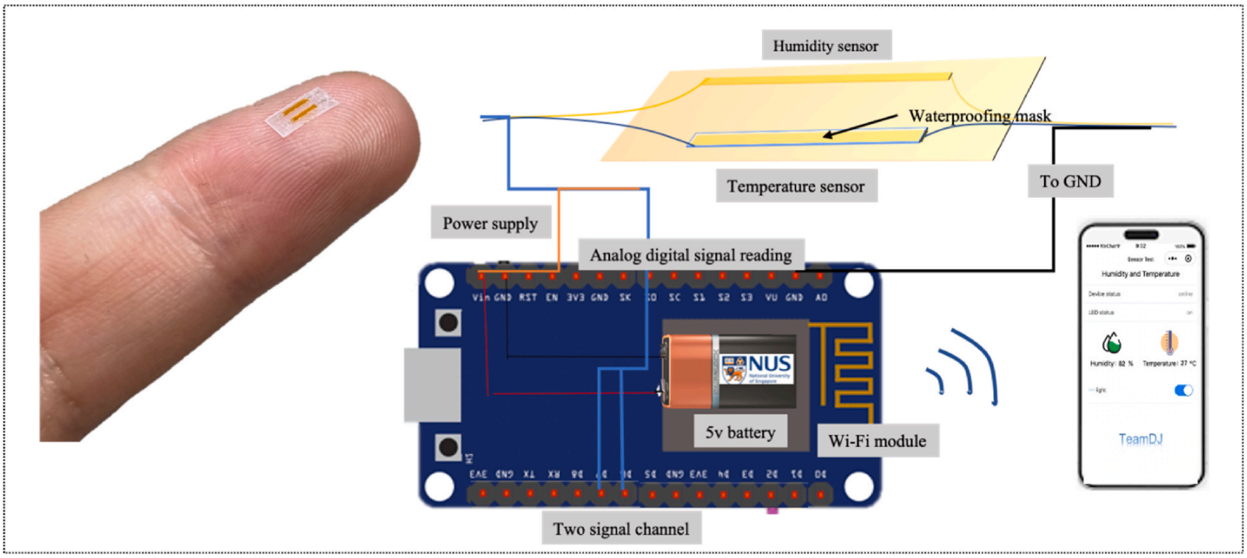

Fig. 7. Schematic and photo of the bimodal sensor.

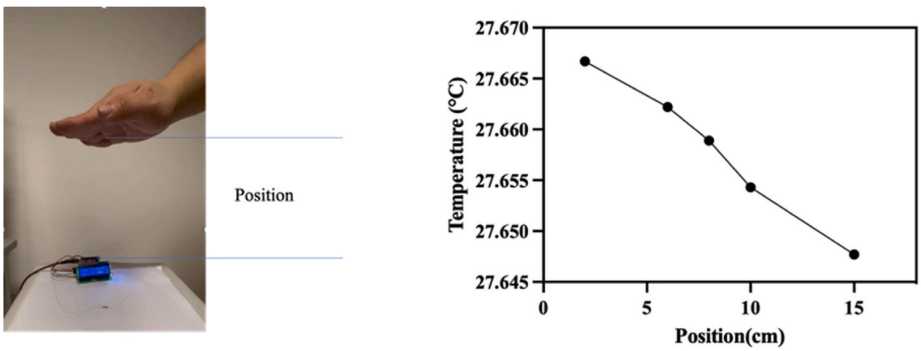

Fig. 8. The response of the sensor to hand movement.

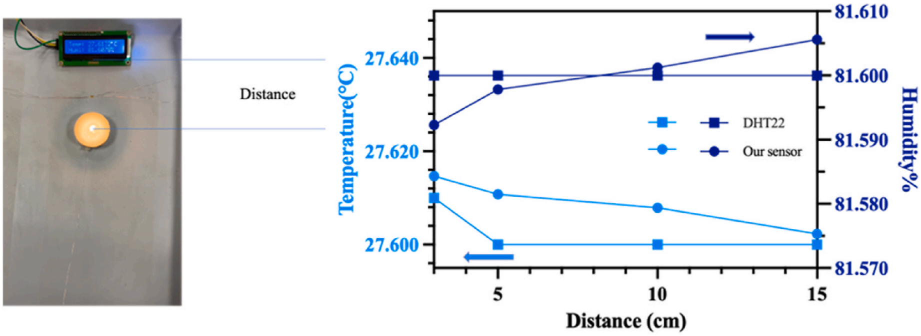

Fig. 9. The response of the sensor to hand movement.

3.6. Breath detection

As previously introduced in the section on the bimodal sensor, it exhibited remarkable sensitivity, capable of detecting changes in ambient temperature as small as 0.004 °C. This exceptional sensitivity allowed us to integrate the sensor with an N95 mask, creating a breath sensor, as depicted in Fig. 10. In this figure, the sensor is positioned on the front arc of an N95 mask and connected to a computer via a small Wi-Fi module as shown in Fig. 10a. The data in Fig. 10b demonstrates the consistency in temperature and humidity variations when the subjects are breathing.

Upon further comparison with other existing works [44,45], it becomes evident that our bimodal sensor performed comparably to current commercial breath sensors. Furthermore, the high sensitivity enabled the sensor strip to be placed on breath masks rather than on the upper lip. This adaptation made it possible to utilize breath sensors on patients requiring oxygen support, while avoiding potential skin irritation often associated with conventional wearable sensors that need skin contact [47].

4. Conclusion

In this work, we have presented a novel stretchable temperature and humidity sensor based on a composite made with elastomer and ionic liquid. The sensor shows high transparency, flexibility, and ionic conductivity, as well as excellent stability and self-adhesiveness. The sensor can monitor both temperature and humidity changes with excellent sensitivity higher than most existing works at around room temperature, and thus is applicable in room temperature sensing and breath sensing. We believe that this polymer has great potential in both academic and applicational uses.

Consent of participants

All participants in this study gave their informed consent to participate in the study and to have their research work published in Heliyon journal. The purpose, description, benefits, risks, confidentiality, and data protection measures of the study were explained to them.

The consenter and consentees' names and contacts are as follows: Consenter: Xian Tongfeng, Email: E0260163@u.nus.edu Consentee: Xu Xin, Email: xin.xu@u.nus.edu Liu Weilin, Email: weilin.liu@outlook.com Ding Jun, Email: msedingj@nus.edu.sg.

Confirmation of authorship

The authors listed out on the title page hereby confirm that, they are the authors of the article Ultrasensitive Stretchable Bimodal Sensor Based on Novel Elastomer and Ionic Liquid for Temperature and Humidity Detection. And have performed the duties as declared in the editorial system. Confirmer: Xian Tongfeng, Email: E0260163@u.nus.edu Xu Xin, Email: xin.xu@u.nus.edu.

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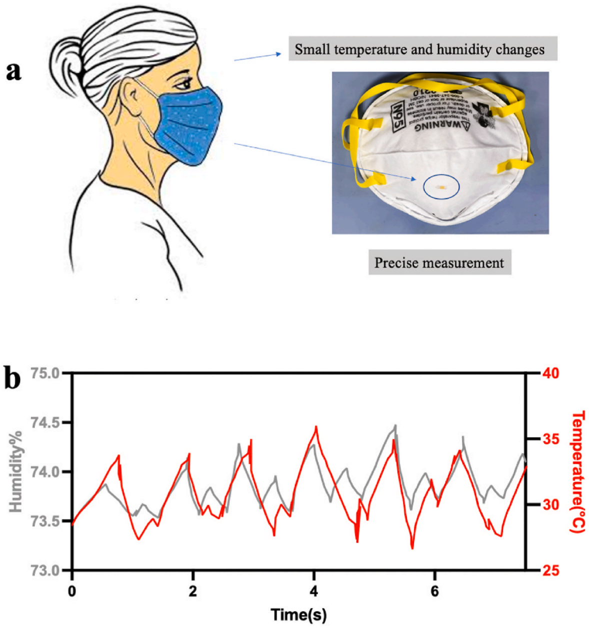

Fig. 10. (a)Schematic and photo of the breath sensor. (b) Breath testing results.

Ethics statement

The authors of this article fully conform to the Publication ethics of the journal.

All participants/patients (or their proxies/legal guardians) provided informed consent to participate in the study.

Review and/or approval by an ethics committee was not needed for this study because there are no ethical dubious actions taken during the course of this study and the writing of this article.

Author notes

Xian Tongfeng is now at the Department of material science and engineering, National University of Singapore.

We have no conflicts of interest to disclose.

Data availability statement

No data was used for the research described in the article.

CRediT authorship contribution statement

Xian Tongfeng: Writing – review & editing. **Xu Xin:** Writing – review & editing, Visualization, Validation, Software, Methodology, Data curation. **Liu Weilin:** Visualization, Validation, Software, Project administration. **Ding Jun:** Writing – review & editing, Supervision, Resources, Project administration, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.heliyon.2024.e25874.

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