

Opto-Electronic Advances

ISSN 2096-4579

CN 51-1781/TN

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Citation: Cui SY, Lu YY, Kong DP, Luo HY, Peng L et al. Laser direct writing of Ga₂O₃/liquid metal-based flexible humidity sensors. *Opto-Electron Adv* 6, 220172(2023).

<https://doi.org/10.29026/oea.2023.220172>

Received: 27 October 2022; Accepted: 22 February 2023; Published online: 25 April 2023

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DOI: [10.29026/oea.2023.220172](https://doi.org/10.29026/oea.2023.220172)

Laser direct writing of Ga₂O₃/liquid metal-based flexible humidity sensors

Songya Cui^{1,2†}, Yuyao Lu^{1†}, Depeng Kong¹, Huayu Luo¹, Liang Peng², Geng Yang¹, Huayong Yang¹ and Kaichen Xu^{1*}

Flexible and wearable humidity sensors play a vital role in daily point-of-care diagnosis and noncontact human-machine interactions. However, achieving a facile and high-speed fabrication approach to realizing flexible humidity sensors remains a challenge. In this work, a wearable capacitive-type Ga₂O₃/liquid metal-based humidity sensor is demonstrated by a one-step laser direct writing technique. Owing to the photothermal effect of laser, the Ga₂O₃-wrapped liquid metal particles can be selectively sintered and converted from insulative to conductive traces with a resistivity of 0.19 Ω·cm, while the untreated regions serve as active sensing layers in response to moisture changes. Under 95% relative humidity, the humidity sensor displays a highly stable performance along with rapid response and recover time. Utilizing these superior properties, the Ga₂O₃/liquid metal-based humidity sensor is able to monitor human respiration rate, as well as skin moisture of the palm under different physiological states for healthcare monitoring.

Keywords: laser direct writing; liquid metal; humidity sensors; flexible electronics; wearable sensors

Cui SY, Lu YY, Kong DP, Luo HY, Peng L et al. Laser direct writing of Ga₂O₃/liquid metal-based flexible humidity sensors. *Opto-Electron Adv* 6, 220172 (2023).

Introduction

Recent studies in emerging flexible humidity sensors have achieved great developments in advanced manufacturing methods^{1–3}, as well as innovative applications including human healthcare detection^{4,5}, plant health management^{6,7} and noncontact human-machine interfaces⁸. Based on different working principles, flexible humidity sensors are mainly categorized into different types including capacitive⁴, resistive⁹, impedance-type¹⁰, voltage-type¹¹ and surface acoustic wave¹². Among them, capacitive-type humidity sensors have gained much attention due to reliable humidity sensing performance, low power consumption and facile structural designs^{13,14}. Generally,

the performance of a capacitive humidity sensor is strongly correlated with the dielectric permittivity of functional nanomaterials between the electrodes¹⁵. Until now, various active materials have been investigated as flexible capacitive humidity sensors, such as carbon materials^{10,16}, cellulose paper¹⁷, metal oxides^{18,19}, metal sulfides^{20,21}, and conductive polymer²². They are typically endowed with high hydrophilicity, large exposed surface areas and rich active sites to interact with water molecules.

Ga₂O₃, as a potential metal oxide with high chemical and thermal stability, has been employed as an active material for humidity sensors^{23,24}. The fabrication techniques to realize Ga₂O₃-based humidity sensors mainly involve chemical vapor deposition (CVD)^{25–27}, thermal

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Received: 27 October 2022; Accepted: 22 February 2023; Published online: 25 April 2023



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treatment²³, and hydrothermal methods²⁸. For instance, Tsai et al. proposed a metal-organic CVD growth method to form β -Ga₂O₃ nanowires as sensitive materials in response to humidity changes²⁵. Furthermore, the response properties of β -Ga₂O₃-GaS humidity sensors fabricated by thermal treatment at the range from 42% to 92% relative humidity (RH) are presented²³. Nevertheless, these traditional methods to develop Ga₂O₃-based humidity sensors usually involve high annealing temperature, complicated fabrication procedures as well as various material systems, hindering their practical applications.

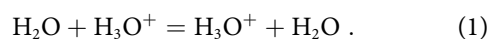
Digital laser direct writing is a rapid and environmental-friendly manufacturing approach to generating functional micro/nano-structures or directly creating sensitive nanomaterials with high precision^{29–32}. Based on laser-matter interactions, via judiciously selecting the appropriate laser processing parameters, a variety of innovative flexible sensors, such as physical, chemical and physiological sensors have been demonstrated^{33–38}. In terms of the flexible humidity sensors by laser processing, the typical strategies usually rely on the laser direct writing of electrodes, followed by the deposition of moisture sensitive nanomaterials, such as carbon or metal sulfides-based materials^{6,8,39,40}. However, electrodes and sensitive materials of these humidity sensors are usually fabricated by different processing technologies with multiple procedures. Therefore, a facile manufacturing approach to developing thin film-based humidity sensor is still required.

In this work, a one-step fabrication strategy based on ultraviolet (UV) laser direct writing to realize Ga₂O₃/liquid metal (LM)-based flexible capacitive humidity sensor is proposed. Owing to the centralized heat of UV laser, interdigital electrodes can be rapidly formed with a well-controlled manner through selectively disrupting the superficial Ga₂O₃ layers surrounding the LM particles. This dexterously converts the insulative Ga₂O₃-wrapped LM (GWLM) particles into conductive paths along with the scanning traces of designed patterns. The unsintered regions with Ga₂O₃ layers between electrodes serve as active sensing nanomaterials in response to humidity variations. The resistivity of conductive patterns can be optimized to as low as 0.19 Ω ·cm. The humidity sensor presents almost no performance degradation after 46 h measurement. The humidity sensor is applied to monitor the respiration and the sweating during exercise. Hence, this work affords a facile fabrication strategy to realize flexible humidity sensors for healthcare monitoring.

Results and discussion

The fabrication process to realize a flexible Ga₂O₃/LM-based humidity sensor is presented in Fig. 1(a), mainly including ultrasonication, spray-coating and laser sintering. The dispersed solution composed of GWLM particles was obtained by ultrasonication of bulk eutectic gallium-indium (eGaIn) alloy in the ethanol (Fig. 1(a-i)). Then, the dispersed solution was spray-coated on a polyimide (PI) substrate to form a uniform layer of GWLM particles (Fig. 1(a-ii)). This homogeneous layer was non-conductive due to the formation of oxidized layers wrapping each eGaIn particle, resulting in the electrical insulation from each other. Significantly, under the laser treatment, the electrical property can be flexibly tuned by the localized heat of focused UV pulse light, which leads to the thermal expansion of GWLM particles, breaks the oxide shell and then enables the conductive core of LM to leak out. This allows the generation of conductive paths (Fig. 1(a-iii) and 1(b-i, b-ii))^{40,41}. Thus, the capacitive humidity sensor was successfully fabricated by this one-step laser processing method. The flexible humidity sensor was composed of two parts, including the uniformly distributed GWLM particles as active sensing materials to the moisture and the conductive electrodes by laser direct writing (Fig. 1(b-i)). Owing to the merits of programmable digital laser patterning, the sizes and configurations of humidity sensors can be dynamically tuned based on controlling laser processing parameters. Here, a typical design of capacitive humidity sensor using interdigital electrodes is employed.

The sensing mechanism of Ga₂O₃/LM-based humidity sensor is similar to the majority of humidity sensors associated with electron transfer processes involving a Grotthuss chain reaction (Fig. 1(b-iii)). At a low RH level, water molecules firstly occupy the hydrophilic sites on the Ga₂O₃ surface to form a chemisorbed layer^{23,24}. Then, additional layers of water molecules are superimposed on the first chemisorbed layer through hydrogen bonding at a high RH level⁴². Because of the large accumulation of water molecules on material surfaces, high-density hydroniums are generated under the applied electrostatic field. Subsequently, the proton-hopping occurs among the hydroniums (H₃O⁺) and water molecules, which can be described as^{8,39,43},



The proton hopping based on the Grotthuss mechanism is responsible for changing the capacitance of

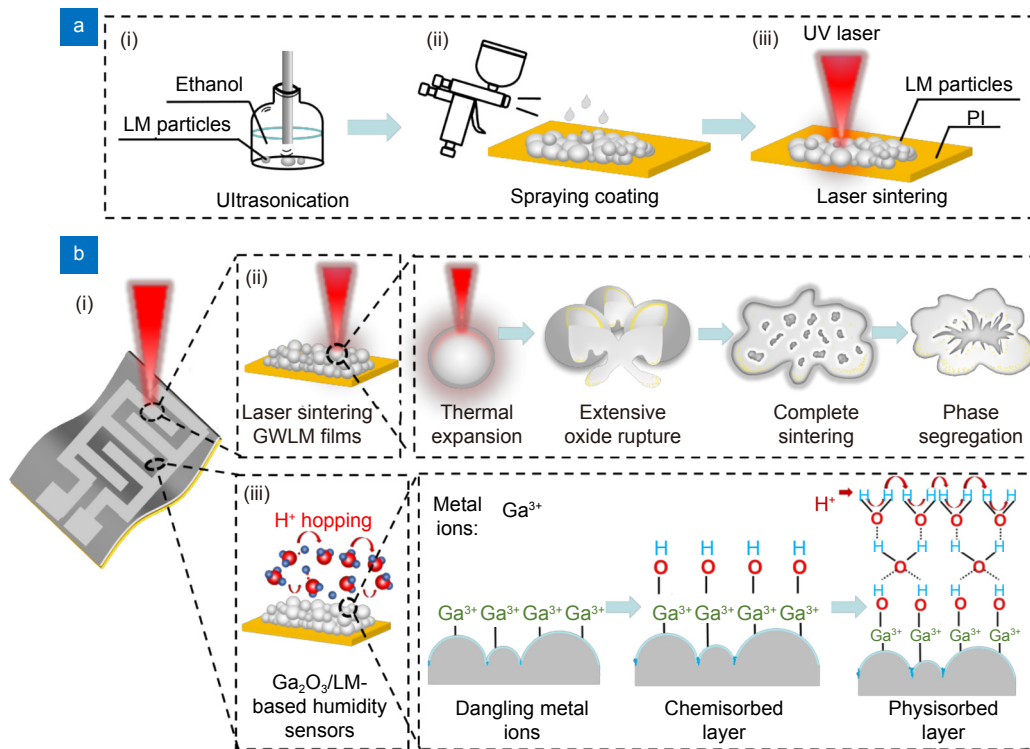


Fig. 1 | Design and fabrication of flexible capacitive humidity sensors. (a) Fabrication processes of flexible $\text{Ga}_2\text{O}_3/\text{LM}$ humidity sensors, including ultrasonication, spraying coating and laser sintering. (b) Schematic of the mechanism to form GWLM films by laser sintering and the sensing mechanism of $\text{Ga}_2\text{O}_3/\text{LM}$ -based humidity sensors.

$\text{Ga}_2\text{O}_3/\text{LM}$ -based humidity sensor. The capacitance (C_{sensor}) can be defined as⁴⁴,

$$C_{\text{sensor}} = \frac{\epsilon_e S}{4\pi k d}, \quad (2)$$

where ϵ_e is dielectric permittivity, S is electrode area, k is electrostatic force constant, d is the distance of parallel plate. In Eq. (2), the C_{sensor} is proportional to ϵ_e when S and d are constant values. The dielectric constant of Ga_2O_3 is about 10.0, while pure water has a larger value of around 78.5^{45,46}. With increasing the RH, the water molecules tend to penetrate into the humidity sensitive layers of GWLM, leading to the increase of capacitance due to the high permittivity of moisture.

Based on the aforementioned principles, this $\text{Ga}_2\text{O}_3/\text{LM}$ -based flexible humidity sensor was fabricated by a pulsed UV laser. To clearly show the surface morphologies of GWLM particles and conductive patterns after laser treatment, scanning electron microscope (SEM) images of corresponding samples were characterized (Fig. 2(a–e)). As observed in Fig. 2(a–e), a fully interconnected liquid metal network was formed, indicating that thermal stress generated by the photo-thermal effect of the laser beam was sufficient to cause extensive rupture of the oxide skin of the particles, allow-

ing liquid cores to flow out and merge into wrinkled continuous structures⁴¹. This implies the formation of conductive paths (Fig. 2(b–d)). To figure out the elemental composition of the sensing materials, the energy-dispersive X-ray spectroscopy (EDX) was also performed (Fig. 2(f)). The elemental mapping results indicated the homogeneous distribution of each element including Ga, In and O in the material system. The ultrasonication and spray-coating of eGaIn on PI film allowed the formation of nonconductive networks with the diameter of most GWLM particles at around 300 nm (Fig. 2(g)). It was found that at a relatively low laser fluence (2.8 J/cm^2), only a few GWLM particles on the surface were sintered into conductive patterns with a resistivity of $\sim 10^5 \text{ } \Omega\text{-cm}$ (Fig. 2(h)). Increasing the laser fluence resulted in the combination of more GWLM particles and then lower resistivity. When the intensity of laser fluence is over 6.8 J/cm^2 , a much lower resistivity at around $0.19 \text{ } \Omega\text{-cm}$ can be achieved. Figure 2(i) shows a conductive LM path with a line width of $48.3 \text{ } \mu\text{m}$. Thus, such a digital laser processing strategy affords to not only pattern LM-based electrodes, but also flexibly tune the conductivity of electrodes. In addition, wettability analysis of this sensor was evaluated by measuring static

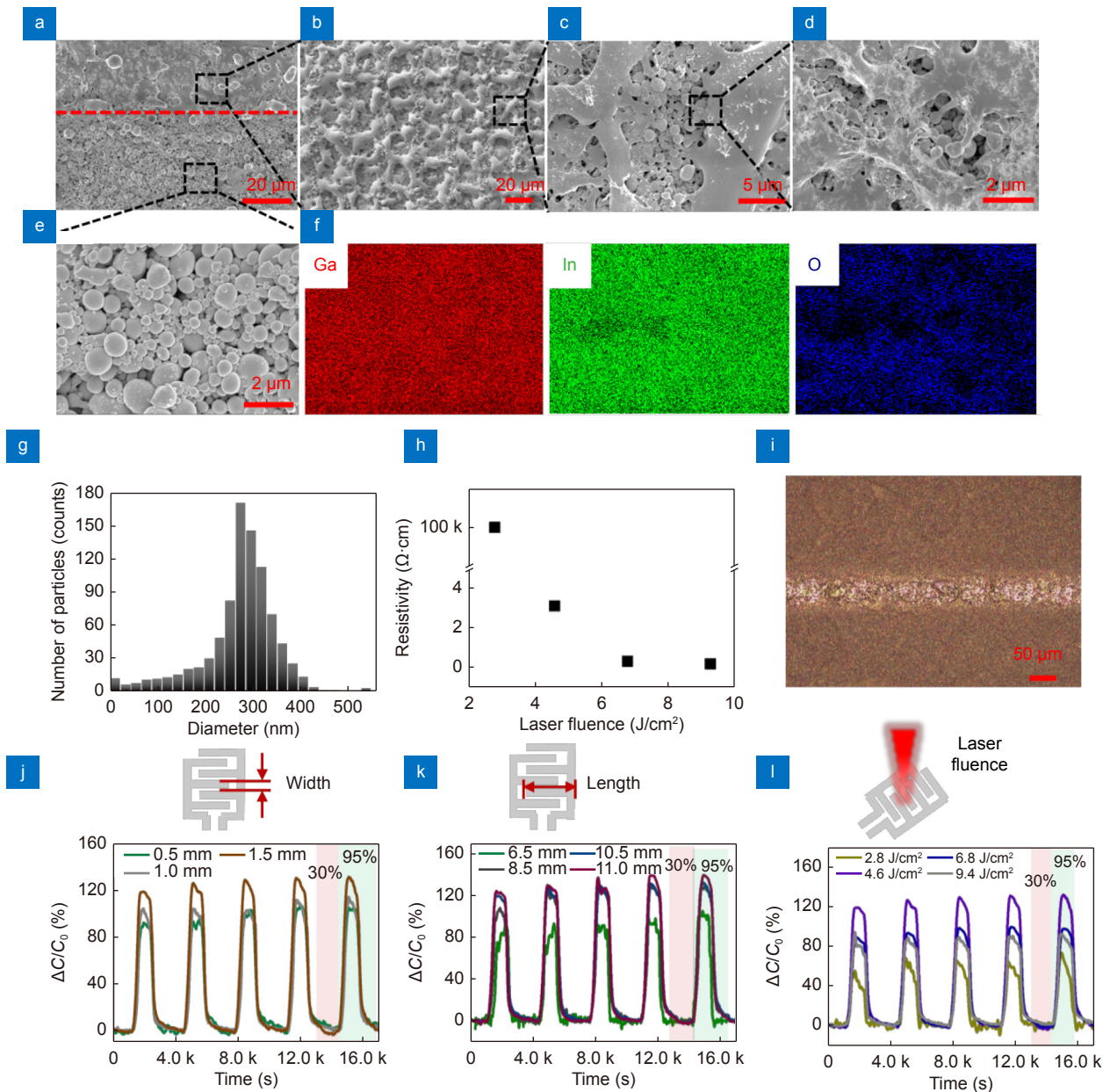


Fig. 2 | Characterizations of flexible humidity sensors. SEM images of GWLM (a–d) with and (a, e) without laser sintering. (f) EDX images of the Ga, In, and O distributions. (g) Histogram of diameter size distribution for the unsintered GWLM particles on the PI film. (h) Resistivity of the laser induced conductive GWLM paths at different laser fluences. (i) The minimum resolution of sintered LM path at a laser fluence of 9.4 J/cm². (j–l) Schematics of Ga₂O₃/LM-based humidity sensors with various fabrication parameters (i.e. widths and lengths of electrodes, UV laser fluence) (top). Cycle measurements of Ga₂O₃/LM-based humidity sensors by periodically varying the humidity from 30% RH to 95% RH (bottom).

contact angles. The water contact angle for unsintered GWLM films was 65° (Fig. S1(a)). In contrast, the contact angle decreases to 19° after sintering GWLM films (Fig. S1(b)). This superior hydrophilic property provides an ideal functional surface to attract water molecules for humidity sensing applications.

To study the humidity effect on the performance of this humidity sensor, the capacitance values were characterized in an oven with controllable temperature and hu-

midity. The humidity sensors with varying interdigital electrodes and laser processing parameters were realized by the one-step laser sintering procedure. During the measurements, the temperature was controlled at a constant value (20 °C). Initially, the cycle tests of Ga₂O₃/LM-based humidity sensors with various widths (*W*) of electrodes were conducted at a humidity range from 30% RH to 95% RH (Fig. 2(j)). It is obvious that the humidity performance shows an increasing trend as the width of

electrodes increases. When the width of electrodes is 1.5 mm, the Ga₂O₃/LM-based humidity sensor exhibits the highest capacitance change of 136.3%. This is probably attributed to the enhanced charge transfer when the gap of adjacent electrodes becomes small (Fig. S2(a, b))⁴². Meanwhile, the performances of humidity sensors with different lengths (*L*) of finger electrodes were investigated (Fig. 2(k)). The Ga₂O₃/LM-based humidity sensor displays the highest capacitance change of up to 142.4% with electrodes length of 11 mm. According to the Eq. (2), the sensing area between active layers and electrodes becomes larger with the longer electrodes, leading to the enhancement of capacitance (Fig. S2(c))⁴⁷. To figure out the relationship between humidity performances and laser fluences, the cyclic performance of Ga₂O₃/LM humidity sensor processed with different laser powers was performed (Fig. 2(l)). It is illustrated that the humidity sensor fabricated at a 4.6 J/cm² laser fluence presents the highest capacitance change of 136.3%. This is due to that at the weaker laser fluence (<4.6 J/cm²), only GWLM particles on the shallow surface were sintered to form the electrode paths (Fig. S2(d)). In addition, at the larger laser fluence (>4.6 J/cm²), the thickness of sintered electrodes become smaller due to the ablation effect. Both cases resulted in the reduction of sensing area between the electrodes and effective functional layer. Therefore, to obtain a humidity sensor with optimal performance, it is significant to select the appropriate parameters including widths and lengths of electrodes, as well as the laser fluence^{48,49}.

In order to compare the performance of humidity devices fabricated by various laser wavelengths, the humidity response of Ga₂O₃/LM sensor sintered by a CO₂ laser was also investigated. The humidity response of the device sintered by the CO₂ laser in Fig. S3 shows relatively poor performance with 47% capacitance change when the RH varied from 30% to 95%. A 355 nm pulsed laser may be a better candidate for laser processing due to that it has relatively smaller thermal effect than CO₂ laser, thus slowing the speed of re-oxidation of the sintered LM electrodes⁵⁰. Therefore, the humidity sensors fabricated by the UV laser are more reliable due to their relatively high sensitivity and reproducibility.

Based on the aforementioned fundamental characterizations, a Ga₂O₃/LM humidity sensor with an electrode width of 1.5 mm and length of 11 mm was created at a laser fluence of 4.6 J/cm² for further investigations. A photo of the Ga₂O₃/LM flexible humidity sensor com-

posed of laser-sintered interdigital electrodes (light grey) and untreated sensing materials (dark grey) is presented in Fig. 3(a). To clearly show the sensitivity of this humidity sensor at different RHs, the capacitance changes of Ga₂O₃/LM humidity sensors were tested from 30% RH to 95% RH. Although the capacitance changes of the humidity sensors present a nonlinear characteristic, the capacitance changes are still clearly captured at a wide variety of RHs. When the RH increases from 30% RH to 95% RH, the humidity sensor displays a capacitance change of ~102.2% (Fig. 3(b)). For the majority of humidity sensors, temperature usually has interference on the output of humidity sensor. To evaluate this cross-talk issue, the capacitance changes of a humidity sensor covered by a piece of gas-proof PET membrane were measured at varied temperatures from 25 °C to 45 °C. The real-time temperatures were monitored by a commercial temperature sensor. Via periodically changing the oven temperature, the Ga₂O₃/LM humidity sensor displays slight fluctuation of capacitance change within 4% (Fig. 3(c)).

Next, to apply the LM-based humidity sensor in practical applications, signal reproducibility is of high importance. Here, 50 cycles' measurement between 30% and 95% RH was conducted (Fig. 3(d, e)). A highly stable cycling behavior was observed at 95% RH for more than 46 h with almost no performance degradation. In addition, four samples with electrodes width of 0.5 mm and length of 11 mm were fabricated by UV laser fluence of 4.6 J/cm². To investigate batch-to-batch reproducibility, cycle measurements of these four different humidity sensors by periodically varying the humidity from 30% RH to 95% RH were tested (Fig. 3(f)). Furthermore, to validate the mechanical bending effect on the humidity sensor, a real-time bending test for the Ga₂O₃/LM-based humidity sensor was performed (Fig. S4). The relatively small variations (<7.4%) under diverse bending radii are probably attributed to the change of the electrode distance and sensing area of humidity sensor induced by the device deformation.

Using the proposed flexible humidity sensors, two important applications were performed. Human breathing monitoring has gained much attention as one of the vital signals for health management. To conduct this, a humidity sensor was attached to a commercial mask on an adult's face (Fig. 4(a)). The humidity response of respiration demonstrates a high repeatability in five measurement periods (Fig. 4(b)). Due to the high moisture induced by the breath, the capacitance change of

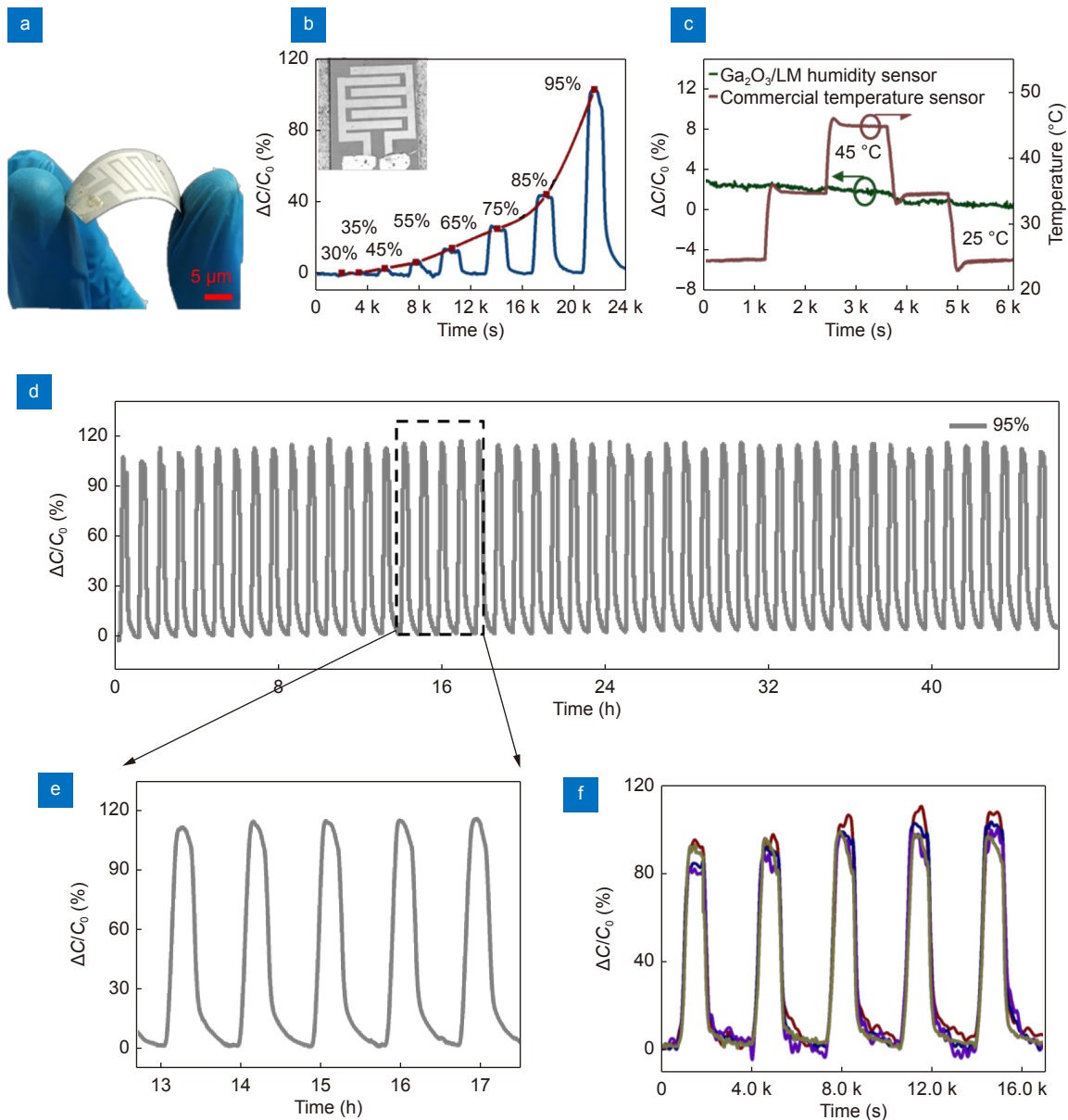


Fig. 3 | (a) Photo of a flexible $\text{Ga}_2\text{O}_3/\text{LM}$ -based humidity sensor. (b) Capacitance change of $\text{Ga}_2\text{O}_3/\text{LM}$ -based humidity sensor at different RHs. The inset shows the image of this sensor. (c) A temperature dependent test of $\text{Ga}_2\text{O}_3/\text{LM}$ -based humidity sensor via varying temperatures from 25 °C to 45 °C. The temperature in the oven was recorded by a commercial thermal sensor. (d, e) Long-term stability measurement (50 cycles) under 95% RH. (f) Cycle measurements of four different batches of humidity sensors by periodically varying the humidity from 30% RH to 95% RH.

193.1% was observed for the respiration monitoring. Furthermore, the humidity performance of long-time breathing monitoring by nose confirms the high sensitivity and fast response of the $\text{Ga}_2\text{O}_3/\text{LM}$ -based humidity sensors (Fig. 4(c–f)). The response and recovery time as two vital humidity parameters to evaluate the response speeds of a humidity device. Notably, this capacitive humidity sensor displays a very fast response time ($t_1 \sim 1.2$ s) according to the respiration test (Fig. 4(c)). This rapid response is probably attributed to enormous numbers of hydrophilic groups inside the Ga_2O_3 particle systems. It

is known that the oxides have high possibility to form hydrogen bonds with the chemically or physically adsorbed water molecules. Such feature provides the Ga_2O_3 nanoparticles with a superior hydrophilic property. The sensing material that is endowed with this property usually presents high sensitivity and fast response due to the effective transfer of protons on the surface occupied by multiple water molecules. Additionally, it took about 1.6 s (t_2) for the device to recover to its original dry condition, indicating that the desorption of water molecules is also fast. These results are comparable to other

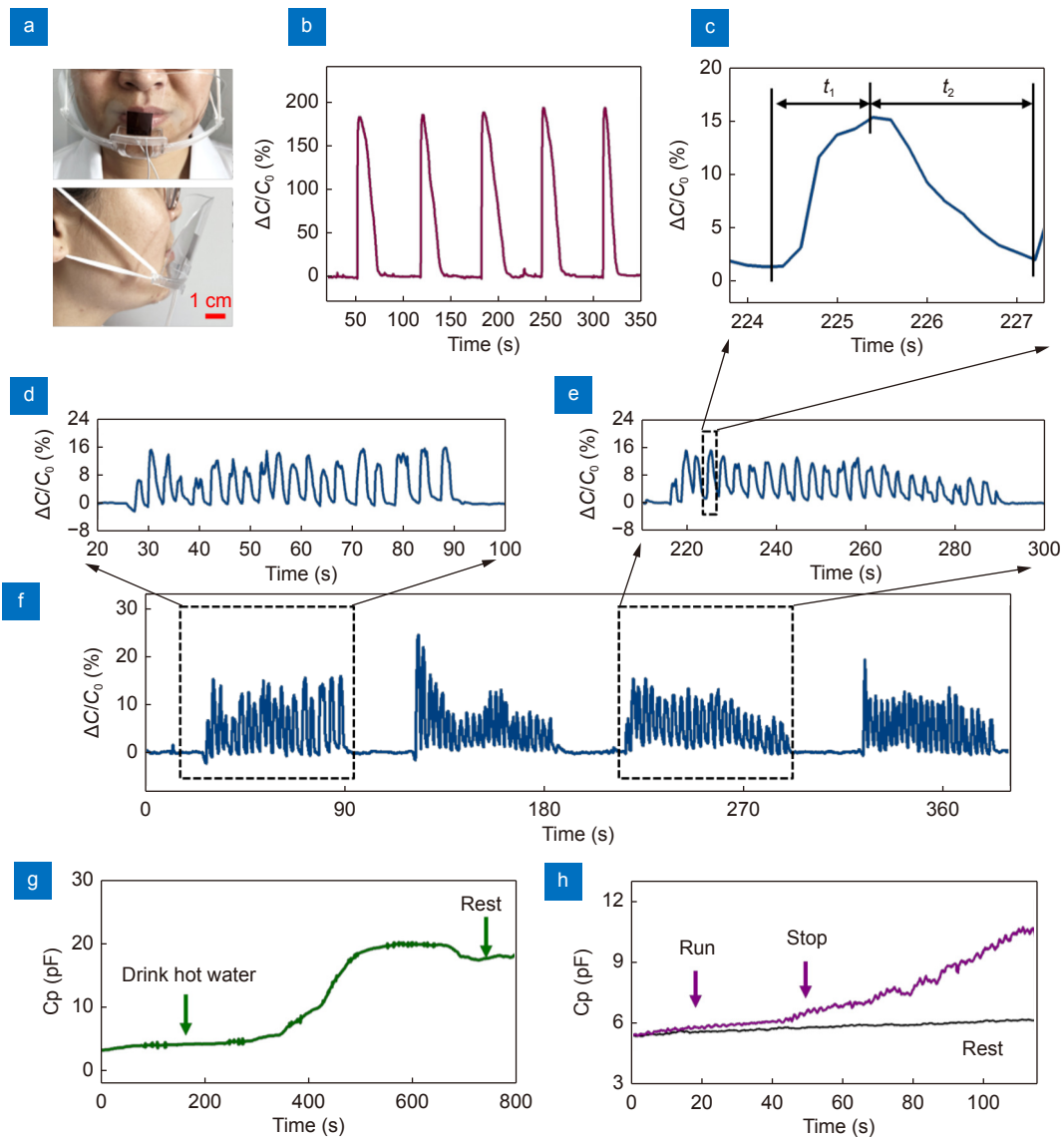


Fig. 4 | (a) Photos of a humidity sensor on a commercial mask worn on the subject's face. (b) Human respiration test of a subject by mouth at a rest state. (c) Response and recovery time of the sensor. (d–f) Real-time monitoring of respiratory rate by nose of a subject at a rest state. Real-time monitoring of palm moisture while (g) drinking hot water and (h) exercising.

thin film-based humidity sensors (Table S1). In addition, the measured breathing rate of this subject was about 18 times/min, which agreed with the respiratory rate (12–20 times/min) of a healthy adult (Fig. 4(d–f))⁵¹.

Another significant application is to detect the sweat evaporation from human skin while drinking hot water and exercising. By attaching the humidity device on the palm of a hand, the moisture condition can be captured during drinking hot water. Generally, the temperature of human body is regulated by heat loss from pores, which is usually accompanied by sweating⁵². A healthy adult usually regulates body temperature by sweating after physical activities. The image of this sensor attached on the hand palm is displayed in Fig. S5. Here, the real-time

humidity variation of a palm during drinking hot water was performed (Fig. 4(g)). At the beginning, the capacitance maintained a stable value at around 3.3 pF. After drinking 150 mL hot water, the capacitance gradually increased. The capacitance change is about 16.7 pF after about 325 s of drinking water and keeps the C_{sensor} at ~ 20.0 pF for around 160 s. This implies that the skin tends to balance the water content in the body and releases the moisture when necessary to decrease the body temperature. After some time, the capacitance of humidity sensor slightly decreased to 18.2 pF, indicating that the body state gradually recovered³⁹. Additionally, the real-time palm moisture under exercising was also monitored (Fig. 4(h)). The water loss process of this adult was

captured at two different physical states (exercising and rest states). At the rest state, the capacitance of humidity sensor shows a relatively stable value (~5.5 pF). Upon running, the capacitance output of humidity sensor gradually increased from 5.4 pF to 10.5 pF, indicating a high moisture level on the palm skin. Even the volunteer stopped running, the capacitance continued increasing, implying the slow recovery of body temperature. In summary, the Ga₂O₃/LM-based humidity sensor can serve as a potential device for real-time healthcare monitoring.

Conclusions

In summary, the Ga₂O₃/LM-based flexible humidity sensors realized by one-step UV laser direct writing approach are studied. The photothermal effect of laser irradiation results in the combination of insulative Ga₂O₃/LM particles, leading to the high conductivity of interdigital electrodes, while the untreated regions are applied as active sensing materials in response to moisture variations. It is found that this humidity sensor presents long-term stability (46 h measurement) with a rapid response (~1.2 s) and recovery time (~1.6 s). This one-step digital laser direct writing approach paves a new way to fabricate flexible and wearable humidity sensors for practical applications towards human health monitoring.

Experimental section

Fabrication of Ga₂O₃/LM-based flexible humidity sensors

The GWLM dispersion was made by mixing eGaIn (75% Ga, 25% In, 200 mg) with ethanol (10 mL), followed by the sonication with a sonicator probe (Dowell, DW-SD28-300B) for 30 min. The solution was simultaneously cooled in an ice-water bath. Prior to spraying, a vortex mixer was applied for vigorous mixing to ensure uniform GWLM dispersion. To obtain uniform GWLM particles on the thin film, a spray equipment was employed consisting of a spray gun (Anest Iwata, W-101) and an air pump (30 L). The GWLM dispersion was sprayed onto the surface of a PI film to form uniform GWLM particle layers, which were naturally dried in the air. To create conductive paths, the GWLM particles were selectively sintered by a nanosecond UV laser (FO-TIA-355-5-30-W, Advanced Optowave, USA) with a wavelength of 355 nm and pulse repetition rate of 20 kHz. The laser fluence was varied from 2.8 to 9.4 J/cm²,

while the scanning speed was fixed at 500 mm/s. In addition, a CO₂ laser (Universal Laser Systems VLS3.50) with a 10.6 μm wavelength was selected as a comparison.

Characterizations of Ga₂O₃/LM-based flexible humidity sensors

Surface morphology of the sensor was characterized by a thermal field emission scanning electron microscope (SEM, Hitachi SU-70 UHR), which was equipped with an energy dispersive X-ray (EDX) detector for the elemental analysis. Resistance measurements of the Ga₂O₃/LM humidity sensors were performed by a digital multimeter (Keysight, 34470A). The relative humidity and temperature responses of the sensor were dynamically measured in a bench-top environment oven (Espec, SH-262). Commercial temperature and humidity sensors were applied to calibrate the temperature and humidity in the oven. The capacitance was measured using an LCR multimeter (Keysight, E4980AL) with a measurement frequency of 100 kHz. The bending tests were conducted by attaching the device onto the curved surfaces with different radii.

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Acknowledgements

This study was supported by the National Natural Science Foundation of China (52105593 and 62271439), STI 2030 —Major Projects (2022ZD0208601) and the “Pioneer” and “Leading Goose” R&D Program of Zhejiang (2023C01051).

Competing interests

The authors declare no competing financial interests.

Supplementary information

Supplementary information is available for this paper at <https://doi.org/10.29026/oea.2023.220172>