Published in partnership with Nanjing Tech University



https://doi.org/10.1038/s41528-025-00446-z

Paper-based flexible electronic devices: processing, integration, and applications

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This paper provides a comprehensive review of the research progress in paper-based flexible electronic devices, focusing on key aspects such as the physical and chemical properties of paper substrates, device structures, fabrication methods for electrodes and active layers, and their diverse applications. The paper also identifies current challenges facing paper-based electronic devices, such as issues related to long-term stability and the optimization of large-scale production processes.

In today's era of digitalization and intelligence, electronic devices, as the key components for acquiring and processing information, are undergoing profound changes and innovations to meet the growing and diversified needs in various fields¹. From environmental monitoring to medical diagnosis, from consumer electronics to industrial automation, the pursuit of electronic devices with high sensitivity, high stability, low cost and easy integration never stops^{2,3}. Paper, as an ancient and widely used material, has gradually become an ideal substrate for the construction of new electronic devices by virtue of its unique physicochemical properties, such as lightweight, porous, biodegradable, flexible, and rich surface chemistry properties^{4–6}. The emerging field of paper-based electronic devices, which not only inherits the advantages of paper, but also shows extraordinary application potential through the organic combination with modern electronic technology, opens up a completely new path for the development of electronic devices.

In the research scope of paper-based electronic devices, in-depth investigation of the physical and chemical properties of the paper substrate is the cornerstone. The precise control of its microstructure and macroscopic properties plays a crucial role in the construction of subsequent electronic functional components. As the core components, the optimization and innovation of the preparation process of electrodes and active layers are directly related to the electrical performance and stability of the devices⁷⁻¹⁰. By skillfully selecting materials and applying advanced preparation techniques, such as printing, deposition, and surface self-assembly method, researchers are committed to realizing the construction of highperformance electrodes and active layers, thus enhancing the overall performance of paper-based electronic devices. Further, based on the functional principle, paper-based electronic devices can be subdivided into various types, including paper-based mechanical sensors, electrochemical sensors, optical sensors, humidity sensors, photodetectors, and supercapacitors^{11,12}. Each type of device plays a unique role in its specific application scenario, such as flexible mechanical sensors for human movement monitoring, electrochemical sensors for environmental contaminant detection and biomarker analysis, optical sensors for food safety and medical diagnostics, and humidity sensors for intelligent packaging and environmental humidity monitoring. Photodetectors are used for information acquisition in optical communication and optical sensing, and supercapacitors for portable energy storage^{13–15}. These diverse applications fully demonstrate the ability of paper electronic devices to solve practical problems in different fields, attracting researchers around the world to optimize the preparation process and performance, which provide more innovative solutions for future smart life, green technology, and precision medicine^{16–18}. In this paper, we will discuss in detail the physical and chemical properties of paper substrates, the structural classification of paper-based electronic devices, the preparation and integration of devices, and applications, aiming to sort out the current research status, analyze the existing challenges, and look forward to the future development direction.

Structure of the paper

Paper-based materials, with their unique physical and chemical structure properties, have become ideal substrates for flexible electronic devices. Specifically, the flexibility of paper-based materials gives flexible electronic devices excellent bending and tensile properties, the lightweight characteristics reduce the overall weight of the device, the porous nature of paper-based materials provides an effective channel for electron transport and material exchange, and the characteristics of the cut and print properties greatly enhance the flexibility and precision of the device design and fabrication¹⁹⁻²¹. In addition, the renewability of paper-based materials is in line with the concept of sustainable development, the easy modification and functionalization provide the possibility of device customization and multifunctionality, and the thermal and chemical stability ensures that the devices operate stably in a variety of complex environments^{22,23}. Together, these characteristics constitute the unique advantages of paper-based materials in the field of flexible electronic devices and lay a solid foundation for their wide application in a variety of application scenarios.

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Physical properties of the paper base

Paper-based materials show remarkable potential in the field of flexible electronics due to their excellent flexibility, low density and lightweight properties, and excellent biocompatibility. Their outstanding flexibility, i.e., the ability to withstand bending, folding, and even twisting without rupture or loss of structural integrity, makes paper-based an ideal substrate for building flexible electronic devices that require bending or folding applications²⁴⁻²⁶. For example, Marina Sala de Medeiros et al.²⁷, in their 2020 study, successfully prepared a self-powered paper-based flexible electronic device that is not only lightweight and flexible, but even has the ability to be folded. Through 500 folding cycle tests, the resistance change rate is less than 2%, which fully verifies the stability and reliability of paperbased materials in maintaining the electrical performance of the device, which is mainly attributed to the excellent flexibility of paper. In addition, the porous structure of paper-based materials facilitates the permeation of gases and liquids, which is particularly critical in the construction of microfluidic devices. Under the capillary force of fiber cavities, paper can be used as a fluid transport channel without the need for an external power device^{28,29}. In a study by Aoxun Liang et al.³⁰ in 2024, a humidity sensor was successfully prepared by impregnating A4 paper with LiCl solution and combining carbon nanotube-carboxymethyl cellulose composite paperbased fiber. The sensor was able to accurately detect humidity over a relative humidity range of 33% to 98%, with a response time of only 500 ms and a linearity of $R^2 = 0.960$. In terms of human health monitoring, this sensor can continuously monitor respiratory rate and amplitude, as well as non-contact sensing of the humidity status of body parts. These properties are achieved due to the loose porous structure of paper, which allows Li⁺ ions to easily adsorb water molecules from the air and ionize to generate H⁺ ions under the action of an electric field.

Another major advantage of paper-based materials is their easy cutting and printing properties, which makes paper-based extremely flexible in the design and fabrication of flexible electronic devices. Xiaoqian Liu et al.³¹, in their study in 2022, prepared an all-paper touch temperature sensor using direct-write technology. This sensor can be freely cut, folded, or reversibly deformed between two-dimensional and three-dimensional configurations without affecting device functionality. Similarly, Huashuo Ma et al.³² developed an editable flexible wood origami electronic device by combining a transparent wood film (TWF) substrate with cellulose-based conductive ink. This device possesses excellent flexibility, high tensile strength, and good electrical conductivity. It can output stable current signals when making origami and printing patterns of various geometrical shapes. These examples further prove the broad application prospects of paper-based materials in the design and fabrication of flexible electronic devices.

Chemical properties of the paper base

Paper-based materials, whose core constituent is cellulose, a natural polymer from plants with unique physicochemical properties, have significant potential in various applications. As the most abundant natural polymer, cellulose is a major structural component of plant cell walls and many microorganisms (e.g., fungi, bacteria, and algae) 33,34. It consists of β -D-glucopyranose units linked by β -(1-4) glycosidic bonds (Fig. 1a, b). The

good thermal and chemical stability, renewability, and biocompatibility exhibited by cellulose opens up a promising prospect for the application of paper-based materials in the field of flexible electronics, especially in medical and biotechnology³⁹⁻⁴³. In the cellulose molecular chain of paper-based materials, the presence of hydroxyl groups and other functional groups provides a wealth of chemically active sites that can react with a variety of chemical substances. This enables the customized adjustment of the chemical properties and functions of the materials. Through functionalization, paper-based materials can combine the inherent advantages of paper with the unique properties of modifiers, thereby expanding their application scope⁴⁴⁻⁴⁷. Specifically, in 2020, Mihut et al.⁴⁸ successfully deposited silver and copper onto the surface of filter paper using a high-vacuum magnetron sputtering system at room temperature. This modification strategy significantly enhanced the antimicrobial properties of the filter paper and effectively inhibited the growth of common wastewater bacteria. Similarly, Guo et al.⁴⁹ developed a new type of colorimetric test paper for metal ion detection through a surface esterification process. Guo et al. used esterification reactions to introduce acetoacetyl groups onto cellulose fibers, generating cellulose acetoacetate (CAA) fibers. Due to the excellent chelating ability of acetoacetyl groups for metal ions, the CAA paper shows obvious color changes when capturing Fe³⁺ and Cu²⁺. In addition, Zhang et al.50 infiltrated metal ions into lignin-containing cellulose nanopaper (LCNP). This method significantly enhanced the water resistance and optical properties of the lignocellulose nanopaper by constructing crosslinking interactions within the fiber network. This approach not only preserves the lightweight, flexibility, and renewability of the paper-based materials but also gives them a wider application potential, especially in applications requiring high water resistance and excellent optical properties.

General layout and processing technology of paperbased devices

Paper-based electronic devices can be divided into two categories based on their functional characteristics: electronic devices and optical devices, each of which exhibits a unique structural configuration and functional principle. As for optical devices, they are classified into colorimetric devices and fluorescent devices based on different optical response mechanisms. Colorimetric devices are based on the difference in light absorption properties of substances, through color changes to reflect the information to be measured or signal detection. Fluorescent devices rely on the fluorescence emission phenomenon of substances, through changes in fluorescence intensity or fluorescence wavelength migration to transmit and detect signals. Considering that electronic devices generally have complex device structures, the following will take electronic devices as an example to introduce the functional composition and device structure of different electronic devices, in order to facilitate the review of device preparation methods.

Electronic device structure

Within the realm of paper-based electronic devices, they can primarily be categorized into resistive, capacitive, photoelectric, field-effect transistor types and diode types. The structure of paper-based resistive electronic

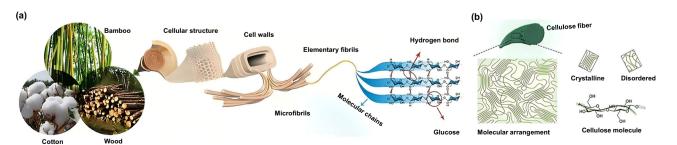


Fig. 1 | Microscopic and chemical structure of paper-based materials. a, b Schematic illustration of hierarchical fibril structure and morphologies of lignocellulose and schematics of the crystalline and disordered regions³⁸.

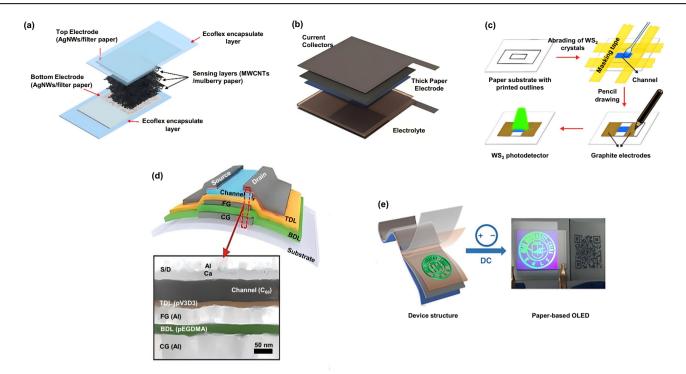


Fig. 2 | The general device structure of paper-based electronic devices. a Schematic illustration of the proposed all-paper-based flexible pressure sensor⁵³. b Schematic representation of thick paper electrode supercapacitor⁵⁵. c Schematic illustration of the fabrication process of paper-based WS₂ photodetectors via

abrading WS $_2$ crystals and penciling graphite electrodes on paper substrates 6 . **d** Schematic illustration of paper-based POFET flash memory consisting of a trilayer gate dielectric structure and artificially-colored TEM image of the flash memory 64 . **e** The Device Structure of Paper-based OLEDs and their Luminescence at 5 V^{71} .

devices typically includes paper-based materials, conductive layers, electrode materials, resistive layers, and encapsulation layers. Paper-based resistive devices usually use paper as the substrate material, and conductive layers are formed on the paper through printing or coating methods. The electrode layer is generally composed of metallic materials or carbon-based materials with good electrical conductivity and stability. The resistive layer is usually achieved through the incorporation or coating of specific materials^{25,51,52}. Paper-based resistive devices respond to external stimuli through changes in resistance. Their basic structure typically involves the integration of conductive materials with a paper substrate⁵³ (Fig. 2a). This structural design allows the device to maintain the flexibility and lightweight nature of paper while achieving precise control over resistance changes^{52,54}.

Paper-based capacitive electronic devices use paper as the substrate, with the core structure being a parallel-plate capacitor configuration. The upper and lower electrode layers are made of flexible conductive materials. Between the two electrode layers is a dielectric layer, which can be selected from polymers (such as polyimide and polyvinyl alcohol) or dielectric structures modified from the paper itself (e.g., impregnated with dielectric liquid or coated with dielectric nanocomposite coatings). Some devices utilize the porous structure of the paper to fill in dielectric materials in order to optimize the dielectric constant. The entire structure is sealed with a polymer encapsulation layer. The electrode leads are connected using flexible conductive traces⁵⁵ (Fig. 2b). The total thickness of the device is usually controlled within 50-150 µm to balance flexibility and capacitive performance^{55,56}. The principle of capacitive electronic devices is mainly based on the charge storage mechanism of capacitors. A capacitor is an element capable of storing electrical charge. Its fundamental principle involves applying a voltage between two conductive plates, causing the accumulation of charge on the plates⁵⁷.

Paper-based photoelectric electronic devices include a core functional layer, electrode layers, and an encapsulation layer. The core functional layer varies depending on the type of device. For example, in paper-based solar cells, a sandwich-like structure is adopted (Fig. 2c), which includes a light

absorption layer (composed of organic semiconductor materials or perovskite materials) and electron/hole transport layers⁵⁸. In paper-based photodetectors, the structure can be planar or vertical, containing a photosensitive layer (made of inorganic semiconductors or organic-inorganic hybrid materials)^{59,60}. Photovoltaic devices are those that convert optical signals into electrical signals through the photoelectric effect. The photoelectric effect refers to the phenomenon where the irradiation of light onto the surface of a metal or semiconductor causes the emission or excitation of surface electrons. These excited electrons are known as photoelectrons. When the energy of a photon exceeds the bandgap width of the material, the photon can excite an electron from the valence band to the conduction band, thereby generating conductive electron-hole pairs⁶¹.

The typical representative of field-effect transistor-type electronic devices is the paper-based organic thin-film transistor (OTFTs). The structure of OTFTs uses paper as the flexible substrate, and from bottom to top, it integrates the gate electrode (such as aluminum, gold, or a conductive polymer layer), the gate insulating layer (using high-dielectric-constant organic/inorganic materials such as polyvinyl alcohol (PVA), polymethyl methacrylate (PMMA), or aluminum oxide (Al₂O₃)), the organic semiconductor layer (formed by solution-processable deposition of p-type or n-type materials such as pentacene or C₆₀), and the source and drain electrodes (composed of metals or conductive polymers) $^{62-64}$ (Fig. 2d). The core mechanism is essentially the dynamic regulation of the conductivity of the organic semiconductor channel by the electric field $^{65-67}$.

The paper-based diode is a general term for various diode devices that use natural paper or modified paper as a flexible substrate. The basic structure of an OLED consists of a thin, transparent electrode layer and another metal cathode, forming a sandwich-like structure. The entire structural layer includes a hole transport layer (HTL), an emissive layer (EL), and an electron transport layer (ETL)^{68,69}. In general, OLEDs need to be fabricated on smooth substrates. Smooth substrates help reduce surface defects and inhomogeneities, thereby improving the luminous efficiency and uniformity of the devices⁷⁰. However, the rough surface structure of



Fig. 3 | Schematic diagram of paper-based electronic device structure.

paper can lead to uneven film coverage and pinhole defects during device fabrication, which in turn affects the transport and recombination efficiency of carriers. Therefore, it is relatively rare to directly fabricate OLEDs on paper surfaces. To successfully fabricate diodes on paper, targeted surface treatment of the paper is required. For example, Teng Pan et al.⁷¹ fabricated a paper-based OLED for high-security anti-counterfeiting applications (Fig. 2e) by using dip-coating treatment and multi-material deposition on commercial paper. This OLED has excellent brightness (maximum brightness of 71,346 cd/m²), efficiency (maximum current efficiency of 64 cd/A), and a half-life of over 4000 h.

Preparation and Integration Methods

Although these devices follow very different physical mechanisms in their functional realization, they show some commonality in structural composition. As shown in Fig. 3. they all contain core components such as paper substrates, electrode materials and active materials. In view of the key role of these basic components in the device performance, the preparation process and integration technology of the electrode part and the active layer are particularly important and constitute the core issues in the research work. Specifically, the selection of electrode materials, structural design, as well as the synthesis of active layer materials, thin film deposition technology, etc., have a decisive impact on the overall performance of the device, so an in-depth study of these preparation and integration technologies is of vital significance to enhance the performance and application scope of paper-based electronic devices. In the selection of electrode materials, carbon family materials (e.g. carbon nanotubes, graphene, carbon black, etc.) are favored due to their excellent electrical conductivity and chemical stability⁷², at the same time, metallic materials (e.g. gold, silver, copper, etc.) are also widely used, in which gold electrodes are known for their excellent chemical stability and electrical conductivity^{73,74}, silver electrodes are competitive due to their relatively low cost, and copper electrodes are of great interest due to their ease of processing⁷⁵. As for the photoelectrically active materials, they cover a wide range of types such as carbon materials and their derivatives, metal oxide semiconductors (MOS), and organic materials⁷⁶. The diverse integration methods of paper-based electronic devices and their structural commonalities provide a broad research space for the selection and preparation techniques of electrode materials and active materials.

The preparation and integration methods of paper-based electronic devices constitute a diversified and technology-intensive field, which mainly includes, but is not limited to, the following high-precision and high-efficiency preparation technologies: screen printing, inkjet printing, electrochemical deposition, vacuum filtration technology, drop casting, and water surface self-assembly method. Each of these technologies has its own unique advantages, and can be flexibly selected and combined according to the specific application requirements and performance indicators of the paper-based electronic devices, in order to achieve high performance, low cost and large-scale device preparation.

Screen printing. Screen printing has become a widely used method for preparing paper electronic devices because of its low cost, good pattern ability and suitable for mass production. The technology is based on precisely transferring ink containing conductive particles to a paper substrate through a screen template, and drying and curing to form conductive patterns. In 2011, Dungchai et al. ⁷⁷ used solid wax-based screen printing technology to

prepare paper-based microfluidic channels. They rubbed solid wax onto a filter paper substrate through a nylon silk mesh and then melted the wax by heating it, allowing it to permeate the paper substrate and form a hydrophobic barrier. Since then, the application of screen printing methods in paper electronic products has become increasingly popular among researchers. (Fig. 4a). Compared with photolithography, the paper-based microfluidic channel prepared by screen printing has a lower background signal, which is convenient for qualitative and semi-quantitative paper-based colorimetric analysis. Thinikan Thongkam⁷⁸ screen-printed an epoxy resin solution with an optimized concentration onto filter paper to form hydrophobic patterns. This process enabled the development of a simple and lowcost 3D paper-based analysis device (3D PAD) for detecting soil available phosphate (Fig. 4b), which exhibits high reproducibility, separability, and stability. The developed PAD shows excellent tolerance to common acids, bases, and organic solvents. Jaruwan Mettakoonpitak et al.79 prepared a paper-based chip using an acetone solution of polycaprolactone (PCL) as a printing ink for the quantification of the important metal Cr3+ and the essential anion Cl⁻. The chip achieved patterning resolution with hydrophilic channels measuring 510 ± 40 µm and hydrophobic edges measuring 490 ± 30 µm. Compared with the other polymer ink, the proposed PCL screen-printed chip achieves approximately 33.3-fold enhancement in the average resolution of hydrophilic channels and hydrophobic edges. Mazzaracchio et al.⁸⁰ prepared a paper-based electrochemical sensor (Fig. 4c) for detecting iron ions in serum. They used graphite conductive ink for screen printing to create the working electrode and counter electrode. The detection limit for iron ions using the Square Wave Voltammetry (SWV) method is as low as 0.05 mg/L. Sun et al.81 used conductive carbon paste as the screenprinting ink to prepare two sets of working electrodes and counter electrodes. Silver/silver chloride ink was used to prepare reference electrodes for constructing a multifunctional self-driven origami-type paper-based microfluidic chip for detecting C-reactive protein (CRP) and prealbumin (PAB) in whole blood (Fig. 4d). The device diverts blood samples separated by plasma separation membranes to the surface of the two sets of working and counter electrodes via capillary force, enabling aptamer-based electrochemical detection of CRP and antibody-based electrochemical detection of PAB. Nair et al.82 prepared paper-based organic electrochemical transistor (OECT) biosensors by screen-printing an OECT on paper with the ability to detect halide ions (Fig. 4e). Paper-based OECTs provide a versatile and costeffective platform for customizable electronic applications and have the potential to be used in disposable devices. This innovative approach offers a new direction for developing recyclable and biodegradable printed OECTs.

Screen-printed paper-based electrodes can realize diverse pattern designs and can be applied to a variety of electrode composites, which helps to prepare electrode systems with different styles and functions, based on which paper-based electrochemical sensors can be constructed with good specific recognition performance, stability and sensitivity. At present, in the field of paper-based electronic devices, exploring new high-performance, green and environmentally friendly screen-printed electrode materials to further improve the accuracy, sensitivity, and stability of paper-based electrodes is the future direction of development. At the same time, the multifunctionality of the electrode materials needs to be considered in order to expand the scope of its application and better meet the application needs.

Inkjet printing. Inkjet printing is another promising technique for the fabrication of paper-based electronic devices, which allows for the fine design of pattern structures by precisely controlling the position and number of ink droplet jets and shows great potential in the preparation of

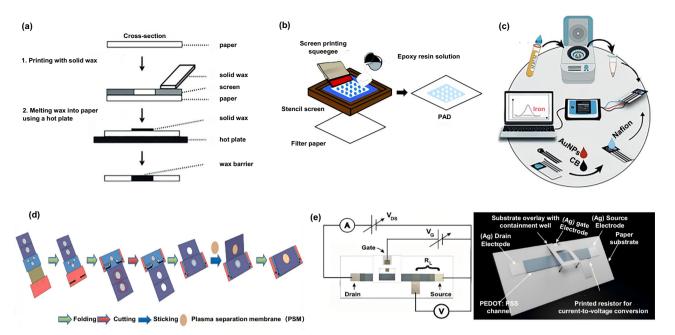


Fig. 4 | Screen printing technology and the fabricated devices. a Constructing paper-based hydrophobic barriers using solid wax-based screen printing technology⁷⁷. b Procedure for making PAD in one step using screen printing method⁷⁸. c Screen-printed paper-based electrochemical sensor for detection of iron

ion in serum⁸⁰. **d** Screen-printed origami paper-based electrochemical chip for detection of C-reactive protein and prealbumin in whole blood⁸¹. **e** Screen-printed OECT based on paper substrate⁸².

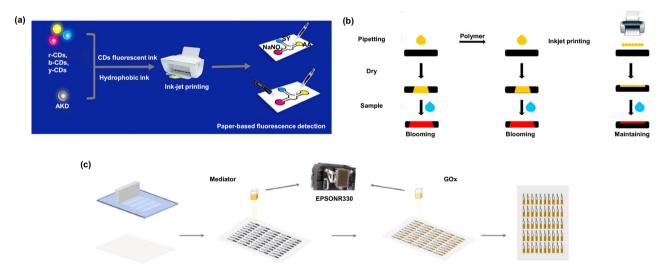


Fig. 5 | Inkjet printing technology and the fabricated devices. a Schematic of preparation of the μPADs based on CDs for fluorescence detection so. b Schematic illustration of pipetting method and inkjet-printing method used for indicator deposition and immobilization c. c. Inkjet printing IPB preparation process diagram so.

paper-based electronic devices ⁸³. Xu et al. ⁸⁴ developed paper-based electronic devices using an alkyl vinyl ketone dimer-heptane solution as inkjet ink to create hydrophilic and hydrophobic patterns on paper in 2010. Since then, inkjet printing has attracted researchers' interests, especially in the preparation of paper-based electronic devices, due to its advantages of low cost, high speed, digital control, ease of operation, and low ink consumption. Kento Maejima et al. ⁸⁵ used an EPSON PX-101 inkjet printer to print UV-curable acrylic ink onto untreated filter paper surfaces, forming microfluidic structural patterns in less than 5 min. After UV curing for 60 s, a hydrophobic barrier was formed, enabling sensitive detection of H₂O. The printed patterns retained their aqueous liquid orientation for at least 6 months at room temperature and for at least 72 h at 50 °C. Yafeng Deng et al. ⁸⁶ developed an all-inkjet printing method by mixing three kinds of fluorescent carbon dots (R-CDs, B-CDs,

Y-CDs) (Fig. 5a). They explored hydrophilic and hydrophobic ink formulations suitable for inkjet printing. The resulting sensor can be used to detect vitamin C (AA), NO₂, and sunset yellow (SY), achieving good visual detection results. Yanqi Li et al.⁸⁷ developed a novel paper-based colorimetric sensor array (Fig. 5b) by inkjet printing pH indicators onto MCE paper using a PEG 400 immobilization system. This method allows for direct pH detection with relatively high resolution due to the precise control of droplet technology and inkjet printing. Compared to other colorimetric-based pH sensors, this sensor array provides a fast, microliter, inexpensive, and accurate method for detecting the pH of normally colorless aqueous solutions. Binghuan Zhang⁸⁸ prepared an inkjet-printed cellulose-based biosensor (IPB) by printing a cellulose-based carbon electrode (PCE) on A4 cellulose paper using an Epson R330 inkjet printer (Fig. 5c). They found that the IPB performed better than a

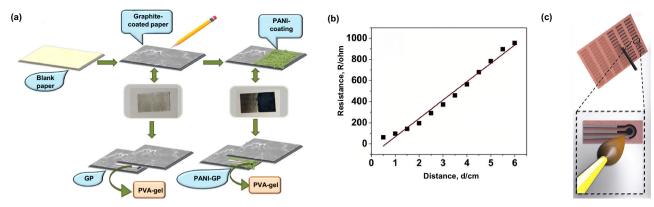


Fig. 6 | **Direct drawing method and the fabricated devices. a** Schematic diagram of preparation of electrodes and fabrication of all-solid-state SCs and (**b**) variation of resistance with distance on the GP electrode surface⁹². **c** The layout of the electrode

(left) and the electrical contacts and reference electrode coated with silver ink are drawn in pencil on the sandpaper substrate (right)⁹³.

drop-coated cellulose paper-based biosensor (DPB). The IPB's response current in the detection range (0–10 mM) was twice that of the DPB. The faster electron transfer rate, flexibility, and repeatability make it an ideal glucose sensor, establishing a new high-performance and biodegradable platform for future single-use glucose detection technologies in daily life.

While the preparation of electronic devices on paper substrates using inkjet printing offers significant advantages, it also faces challenges such as substrate limitations, conductivity issues, and process complexity. For example, different types of substrates may affect print quality and electrode performance. Additionally, a variety of conductive inks, such as silver nanoparticles, graphene, and PEDOT:PSS, have stability and dispersion issues that limit their application. Moreover, inkjet printing requires precise control of ink jet characteristics and post-processing to achieve the desired performance.

Direct drawing method. Preparation of electrode patterns by drawing directly on paper with a pencil is a simple and economical method for paper-based electronic devices. This method is based on the fact that the graphite material in the pencil core has good electrical conductivity, and the electrodes can be formed by directly drawing the desired electrode patterns on the paper. Specifically, the pencil core is mainly composed of a mixture of graphite, clay and wax, of which graphite is the key component for electrical conductivity89. When a pencil is used to draw on paper, graphite particles adhere to the surface of the paper and are embedded in the fiber structure of the paper. As the drawing process proceeds, the graphite particles form continuous conductive paths on the paper 90. In the early days, Li et al.⁹¹ prepared micro-supercapacitors by using medical tape as a flexible and stretchable substrate, using MnO₂ as an active material, and drawing with the pencils, since then, direct pencil drawing on paper has become a common method for preparing paper-based electrodes due to its low cost, simplicity, and fairly good conductivity obtained. For example, Sabina Yeasmin et al. 92 prepared an all-solidstate supercapacitor (SCs) by applying 8B pencil on A4 paper as electrodes and modified paper-based graphite (GP) electrodes with polyaniline nanofibers (Fig. 6a). This SCs now exhibits excellent electrochemical performance and linear variation of the surface resistance of the graphite layer drawn by the pencil with distance (Fig. 6b). Danielly S. Rocha et al. 93 developed a new electrochemical paper-based analytical device (ePAD) (Fig. 6c) by rubbing a graphite pencil directly against the surface of sandpaper for the square-wave voltametric measurement of midazolam maleate used as a "date-rape drug" in beverages. This ePAD provides high accuracy for midazolam maleate, used as a "date rape drug" in beverages. At 95% confidence level, there was no statistical difference from the data recorded by High Performance Liquid Chromatography (HPLC).

In conclusion, the direct mapping method has significant advantages. It is low cost, easy to handle, environmentally degradable, and requires no complex equipment or processes⁹⁴. Moreover, due to the flexibility of paper, the electrodes are also flexible and suitable for the preparation of various flexible electronic devices. However, the roughness and non-uniformity of the paper surface may lead to differences in the conductivity of the electrodes. Furthermore, the limited precision of pencil drawing may not meet the preparation requirements for certain high-precision electrodes. Overall, the pencil-on-paper drawing method for electrode preparation is a simple and practical method for a variety of low-cost, low-precision electrode preparation requirements.

Electrochemical deposition. Electrochemical deposition is a technique in which a material is deposited on a conductive substrate by an electrochemical reaction to prepare a film or coating⁹⁵. Electrochemical deposition usually involves an electrolytic cell containing an electrolyte solution, an anode and a cathode. In the presence of an electric current, ions in the electrolyte solution undergo a redox reaction on the surface of the electrodes, resulting in the deposition of the desired material formed⁹⁶. In the case of paper substrates, they need to be made electrically conductive first, then they can be used as cathodes or anodes for electrochemical deposition⁹⁷. Compared to other electrode preparation methods, the electrochemical deposition method does not require expensive equipment and complex processes, and the production cost is low. Moreover, the electrochemical deposition method is able to control the deposition thickness to form a good bond between the deposited layer and the conductor, which helps to improve the performance of the electrode 98,99. However, electrodeposition does face two key issues: the dependence of electrodeposition on the conductive substrate and the controllable electrodeposition area. To solve this problem, one approach is to first electrodeposit the desired metal pattern on a conductive substrate, incorporate photolithography, and then transfer it to an insulating substrate 100,101. Another approach is to first form patterned conductive channels on the insulating substrate by chemical plating, and then electrodeposit them to form a continuous metal film 102,103. However, these methods either increase the complexity of the process or involve environmentally unfriendly chemicals, which limits their application. On this basis, Xuanzhang Li et al. 104 proposed a new transverse electrodeposition method called "CCB edge-guided metal transverse electrodeposition (EG-MLED)". In this method, an electroactivated carbon patterned layer is sprayed onto the substrate surface to effectively guide the rapid deposition of copper ions on the insulating substrate (Fig. 7a). This EG-MLED method eliminates the dependence of conventional electroplating on conductive substrates

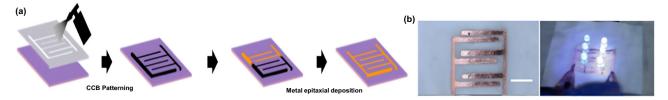


Fig. 7 | Electrochemical deposition method and the fabricated device. a Schematic of the patterned metal film prepared by EG-MLED on CCB template. b Paper-based LED device prepared by EG-MLED¹⁰⁴.

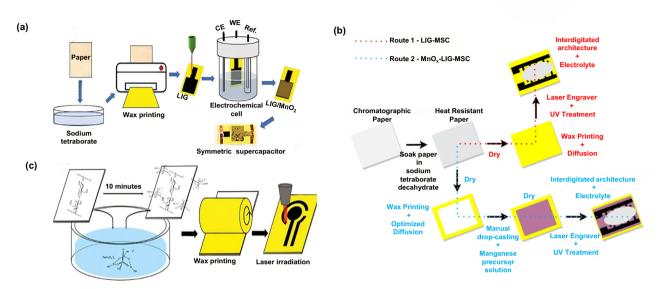


Fig. 8 | Laser induction method and the fabricated devices. a Scheme for the manufacture of LIG on paper and the electrochemical deposition of MnO₂¹¹⁰. b LIG-MSC and MnO_x-LIG-MSC fabrication process¹¹¹. c Chemical treatment imposed on paper substrates, with sodium borate acting as fire-retardant¹¹².

and is particularly suitable for the preparation of metal films on insulating substrates (cellulose paper) with complex surface structures (Fig. 7b), and paper-based light-emitting diode and photodetector devices prepared using this method demonstrate potential applications in flexible electronics.

Laser induction. Laser-induced graphene (LIG) is a simple one-step low-cost method for producing graphene and designing devices directly on carbon-based substrates. Under laser irradiation, the carbonhydrogen bonds on the surface are cleaved and SP³ carbon atoms are converted to SP² carbon atoms through a photothermal conversion process. As a result, LIG consists of a network of conductive porous interconnections with a high surface area for enhanced electrochemical performance and charge transport. In addition, LIG is formed directly on the substrate, simplifying the device fabrication process as it can be used as both a collector and an active material 105-108. In the early days, Zang et al. 109 produced conductive molybdenum carbury-graphene (MCG) composites directly on the paper substrate, and since then, the generation of LIG on the surface of the paper and the simultaneous realization of accurate cutting of the paper and clever construction of the device have been extensively studied. For example, Maykel dos Santos Klem et al. 110 prepared MSC (micro-supercapacitors) with high energy density by preparing LIG electrodes on a wax-coated paper substrate and electrodepositing MnO₂ on the LIG (Fig. 8a). Upon laser heating, paper cellulose fibers were transformed into highly porous LIG structures, increasing the area available for MnO₂ deposition, and MSCs with a capacitance of 86.9 mF cm⁻² could be produced by tuning the laser parameters and deposition time. Rodrigo Abreu et al. 111 laser processed paper fibers embedded with MnO_x precursor converted to graphene to form MnO_x-LIG to obtain MnO_x-LIG-MSC (Fig. 8b). Since MnO_x can promote pseudo-capacitive charge storage to increase the overall capacitance, and LIG has unique porous and conductive properties that provide a favorable substrate for efficient charge transport, the obtained $\rm MnO_{\it x}$ - LIG-MSCs have high specific capacitance, high energy density, and good cycling and electrochemical stability. Tomá s Pinheiro et al. 112 achieved photothermal conversion to porous laser-induced graphene (LIG) on paper through appropriate chemical treatment using sodium tetraborate as a flame retardant (Fig. 8c). In addition, by manipulating the operating parameters of the laser, they have created a high-throughput, easy-to-adapt disposable electrochemical sensor, a low-cost material and manufacturing technique with potential applications in bioelectronics and wearable devices.

In conclusion, applications in the field of paper-based microfluidics and wearable devices are promising, but the requirements for laser equipment are relatively stringent, the operation process is relatively complex, and precise control of the laser parameters is required to ensure that the quality and homogeneity of the active layer are optimized ^{113–116}. In addition, during the laser induction process, some by-products may be generated, which may adversely affect the performance of the active layer ^{117,118}. Therefore, in practical applications, it is necessary to further optimize the laser parameters and improve the subsequent treatment process to minimize the influence of by-products and ensure the performance and stability of the devices.

Drop casting. Drop casting is a simple method for preparing paper-based electronic device films without the need for complex equipment. The basic principles include the preparation of liquid precursors, the formation and deposition of droplets, and the curing and forming of films. Each of these steps in the process has a decisive impact on the quality and performance of the final film. The drop-casting method can realize high-precision and high-uniformity film preparation by precisely

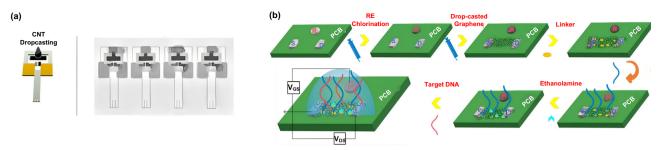


Fig. 9 | The devices fabricated by drop casting. a Drop-casting of suspensions on the sensor electrodes¹¹⁹. b PNA-DNA hybridization detection employing an electrolytegated graphene field-effect transistor¹²⁰.

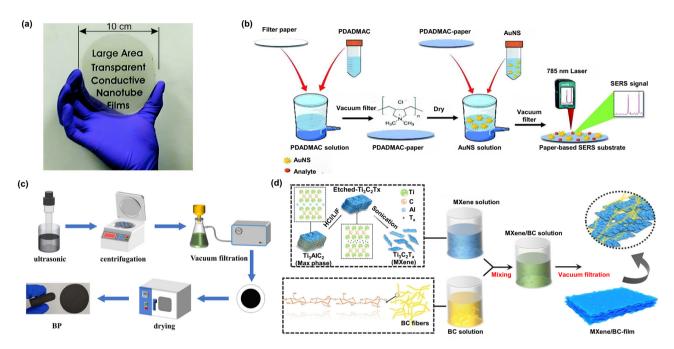


Fig. 10 | The devices fabricated by vacuum filtration. a Preparation and Characterization of CNT film by vacuum filtration 121. b Schematic showing each step of plasmonic paper preparation process 122. c Schematic diagram of BP preparation process 123. d Schematic of MXene/BC film 124.

controlling the volume and deposition position of droplets, and is especially suitable for the fabrication of small-area and complexstructure films, with higher flexibility and process controllability. In recent years, the drop-casting method has been favored by researchers due to its simplicity and speed. For example, Christopher J. Valentine et al.¹¹⁹ drop-cast CNT suspensions into laser-cut electrode areas in multiple steps to build a series of different conductive networks in paper (Fig. 9a). By defining this geometry, CNT suspensions can be simply drop-cast onto paper without the need for additional patterning methods (e.g., inkjet printing or screen printing) to define the electrode regions. Papamatthaiou et al. 220 drop-cast graphene ink to form a transistor channel, and immobilized a PNA probe onto the graphene channel to prepare an electrolyte-gated field effect transistor (FET) biosensors (Fig. 9b) for selective detection of complementary DNA sequences. The results showed that FET sensors have the potential to be effortlessly integrated into Lab-on-PCB diagnostic platforms, promising to further reduce the time for DNA amplification and detection modules.

In summary, drop-coating is easy to manipulate but can lead to inhomogeneous deposition and limited control over the thickness of the active layer. Drip-casting method for preparing the active layer on paper substrate has the advantages of easy operation and lower cost, but it also has the disadvantages of difficulty in controling the film quality, weak bonding

between the active layer and the paper substrate, and difficulty in preparing large-area homogeneous films.

Vacuum filtration. Vacuum filtration is an effective method for preparing thin films for paper-based electronic devices, which is easy to operate and master, and can prepare uniform and dense films. It is suitable for the preparation of a variety of electronic materials, such as conductive polymers, nanoparticles and so on. The basic principle is to use a vacuum pump to generate negative pressure, so that the solution or dispersion through the filter membrane, the solvent in the solution or dispersed media pumped out, while the solute or dispersed material is deposited on the filter membrane to form a thin film. In the preparation of paper-based electronic devices, this method is commonly used in such as conductive polymers, nanoparticles uniformly deposited on the paper substrate to form a film with a specific function. In the early stage, Wu Z et al.¹²¹ deposited carbon nanotubes on the filter membrane by vacuum filtration method to prepare carbon nanotube films, which had excellent electrical conductivity and light transmission (Fig. 10a). Since then, the vacuum filtration method has been increasingly applied to the preparation of paper-based electronic devices. For example, Kanyawan Ponlamuangdee et al.¹²² modified ordinary laboratory filter paper with the cationic poly (diallyldimethylammonium chloride) (PDADMAC) and used a simple vacuum filtration method to develop a paper-based Surface



Fig. 11 | Surface self-assembly method. a Diagram of the preparation of bionic structured PCPM¹²⁵. b Schematic of the structure of GCEF fabricated at the water/air interface¹²⁶. c Illustration showing the preparation of graphene films as the sensing layer of a-G@E sensor¹²⁷.

enhanced Raman scattering (SERS) substrate (Fig. 10b) for the detection of pesticides, including paraguat, diquat, thiophanate and parathion. This substrate exhibited excellent inter-spot reproducibility with a relative standard deviation (RSD) of 5.03% and low detection limits of 0.51 µM (0.13 ppm) for paraguat and 0.38 µM (0.09 ppm) for thiophanate. Ru Chen et al. 123 prepared composite films based on whisker-carbon nanotubes (w-CNTs) by vacuum filtration using Triton-100 as a surfactant (Fig. 10c). The films obtained by this method have uniform and dense surfaces, and w-CNT exhibits excellent electrothermal properties due to the more complete and higher crystallinity of the w-CNT structure. Tuoyi Su et al. 124 prepared MXene (Ti₃C₂T_x)/Bacterial cellulose (BC) film by mixing MXene and BC with a mass ratio of 1:4 and vacuum filtration (Fig. 10d). MXene can be used as a binder to provide filmforming ability and improve the conductive ability of the film due to its multiple chemical bonds. In addition, BC expands the layer spacing of MXene nanosheets and increases the mechanical strength of the material by combining adjacent nanosheets, so that the film exhibits great flexibility and foldability.

In summary, vacuum filtration for preparing active layers on paper substrates offers advantages such as high efficiency, scalability, high material utilization, and controllable preparation processes. However, there is a risk of clogging, which can affect filtration efficiency and layer quality, potentially leading to unstable performance and incomplete filtration. Additionally, the adhesion between the active layer prepared by vacuum filtration and the paper substrate may be weak, necessitating subsequent treatment to enhance bonding. Therefore, this technology presents both potential and challenges. Optimizing filtration conditions and improving processes can further enhance the quality and performance of the active layer, providing strong support for the development of paper-based electronic devices.

Surface self-assembly method. The water surface self-assembly method involves two steps, first, self-assembly on the water surface using the intermolecular interaction forces of the nano-dispersion to form an ordered layer of nano-film, then, this layer of already selfassembled nano-film is transferred from the initial water surface to another target substrate for further application or treatment of the film. This approach combines the advantages of both self-assembly and film transfer techniques and provides a new way to prepare high-quality, ordered-structured films. However, the self-assembly process of nanofilms on aqueous interfaces still faces several challenges and dilemmas that hinder the successful preparation of desirable films. Therefore, it is crucial to optimize and refine the preparation process of self-assembled thin films. In recent years, extensive and in-depth research has been conducted in academia on preparing thin films via self-assembly strategies using water surfaces as templates. This research aims to overcome existing limitations and promote technological advancement and application expansion in this field. For example, Shan Li et al. 125 first employed an ethanol-assisted carbon material suspension, which was skillfully applied on top of the water surface by the spraying technique. They also adjusted the position of the sprayer in order to ensure the homogeneity of the resulting film. Immediately thereafter, the siphoning effect of the microporous sponge was utilized to effectively remove the water, a process that not only significantly reduced the area of the film, but also facilitated the tight alignment of the film's internal structure. Through the above steps, they successfully prepared a uniformly spread carbon nanotube (CNT) film, which was then further transferred onto a polydimethylsiloxane (PDMS) substrate (Fig. 11a). Notably, because the film was prepared in a free-support environment, it is highly transferable and can be easily transferred to other substrate materials such as paper without compromising its original properties. This discovery not only broadens the application scope of self-assembled films, but also provides new ideas and strategies for the development of highperformance and multifunctional film materials. Wei Zhou et al. 126 prepared graphene/carbon sphere films (GCH) by self-assembling on the water surface. They then sprayed Ecoflex on them to increase the mechanical flexibility of the films to form graphene/carbon sphere/ Ecoflex elastic films (GCEF) (Fig. 11b). Finally, leveraging the freesupporting nature of the liquid substrate, they easily transferred the GCEF to a PET substrate. Similarly, the GCEF prepared by this method can also be transferred to a paper substrate while retaining its original properties, due to its free-supporting nature. Shuai Wang et al. 127 prepared a graphene film (GF) by spraying a graphene-ethanol dispersion onto a water surface using a self-assembly method. The GF was then transferred onto an Ecoflex film/PET substrate using a lift-andtransfer method. Subsequently, an additional Ecoflex layer was spincoated to create a sandwich-structured a-G@E sensor (Fig. 11c). The free-standing film prepared by this method not only enhances the mechanical flexibility of the film but also allows for multiple retransfers several times to prepare films with different thicknesses. Furthermore, it can be transferred to paper substrates.

Physical vapor deposition (PVD) method. When fabricating paperbased electronic devices using Physical Vapor Deposition (PVD), materials are first vaporized into atoms or molecules through evaporation, sputtering, or other methods, and then transported to the surface of the paper substrate to deposit a film in a vacuum environment¹²⁸. The paper substrate needs to undergo surface pre-treatment (e.g., coating with PMMA) to fill pores and reduce roughness to adapt to the PVD process. This method faces the challenge of insufficient thermal stability of paper, as paper typically decomposes at temperatures below 200 °C, while the high temperatures in PVD processes can lead to carbonization or deformation of the paper. Additionally, the rough surface of the paper substrate can easily cause defects such as pinholes and uneven coverage in the deposited film, leading to device short-circuiting or performance degradation. Therefore, research in this direction is limited by process compatibility, and there are relatively few published papers. However, in recent years, there have still been representative studies focusing on this direction. For example, Ihalainen et al. 129 prepared ultrathin gold film electrodes on a nanostructured latex-coated paper substrate using PVD



Fig. 12 | Physical vapor deposition method and the performance of the fabricated devices. a A schematic illustration of the structure of the multi-layer coated paper used as the coating substrate for nanostructured two-component latex coating. b Capacitance vs frequency log-log curves for the bare and coated UTGF electrodes¹²⁹.

(Fig. 12a). The device exhibits conductivity comparable to that of bulk gold (with a 6 nm thick gold film), and shows good mechanical stability, maintaining performance unchanged through multiple bending and repeated folding-unfolding cycles. Moreover, the electrode demonstrates excellent stability in electrochemical experiments (Fig. 12b), making it suitable for the development of paper-based electrochemical platforms, such as for biosensing applications.

Applications of paper-based electronic devices

In the research field of paper-based electronic devices, a series of innovative applications are gradually revealing their promising potential. These devices mainly include paper-based mechanical sensors for monitoring and responding to external mechanical stress changes 130, paper-based electrochemical sensors for detecting the presence or concentration of specific chemicals through electrochemical principles¹³¹, paper-based optical sensors, which utilize optical effects to achieve sensing and conversion of light signals¹³², paper-based humidity sensors, which measure and monitor environmental humidity changes¹³³, paper-based photodetectors, which combine optical and electronic properties for detecting optical signals and converting them into electrical signals¹³⁴, and paper-based supercapacitors, which serve as highly efficient energy storage elements to provide the necessary electrical support for paper-based electronic systems¹³⁵. These diverse paper-based electronic devices not only expand the functional boundaries of paper beyond its traditional role as an information carrier, but also bring revolutionary technological advances to fields such as wearable devices, environmental monitoring, and healthcare¹³⁶.

Paper-based mechanical sensors

In the field of flexible electronics and wearable technologies, paper-based mechanical sensors have garnered extensive attention due to their unique advantages. These sensors come in a variety of types, mainly including paper-based resistive sensors, paper-based capacitive sensors, and paper-based piezoelectric sensors. With the advancement of material science and micro/nano-fabrication technologies, the performance and application scope of paper-based mechanical sensors are expected to be further expanded, offering more possibilities for the design of future intelligent systems.

Paper-based resistive stress sensors. In recent years, piezoresistive flexible devices that directly convert pressure into electrical resistance signals have become a research focus due to their excellent mechanical properties, simple device structure, and ease of obtaining output signals. In order to significantly enhance the sensitivity of the sensors, Chao Ji et al. ¹³⁷ skillfully combined the permeability of a paper substrate with the electrical conductivity of graphene, and successfully prepared graphene-based piezoresistive pressure sensors by utilizing multilayer structured cellulose paper as a substrate (Fig. 13a). Notably their seven-layer cellulose paper sensor designed by them demonstrated an excellent sensitivity, with specific values as high as 44 kPa⁻¹ (Fig. 13b), which opens up new avenues for sensor sensitivity enhancement. Xiaoqian Liu et al. ³¹

developed a new foldable touch pressure sensor by coating carbon black ink on cellulose paper (Fig. 13c), which can be freely shaped and reversibly deformed between 2D and 3D configurations without losing functionality, Owing to the significant resistance variation and air-gap effect under compression, the sensor exhibits a high sensitivity of 2.36 kPa⁻¹, along with fast response and recovery characteristics (Fig. 13d). Muhammad Hasnain et al.¹³⁸ created a strain sensor by drawing directly on cellulose paper with a pencil, and SEM showed that the micro-nano structure of graphite penetrated into the void of the fiber frame of cellulose paper, significantly improving the overall electrical conductivity of the film (Fig. 13e). The sensor exhibits good stability and high sensitivity of 122,701 kPa⁻¹ (Fig. 13f), as well as excellent performance at 3.23% bending strain, with response and recovery times of 0.59 s and 0.69 s, respectively (Fig. 13g).

To further improve the sensitivity, Bowen Zheng et al. 139 prepared an ultra-high sensitivity resistive pressure sensor with a sensitivity of 1014 kPa⁻¹ (Fig. 13h) and a wide sensing range of 300 kPa (Fig. 13i) by squeegee coating and screen printing techniques. This sensor was used to effectively monitor various physiological signals, including pulse, body acoustics, joint flexion, and finger tapping. Rajat Subhra Karmakar et al. 140 developed a pressure sensor by combining and folding conductive paper substrates face-to-face, optimized for wearable devices for monitoring teething disorders and neck posture. The average surface roughness of the graphene ink film was 0.634 µm, which created multiple electrical contacts at the interface (Fig. 13j), the sensor achieved a high sensitivity of 3.75 kPa⁻¹ in the 0-0.05 kPa range (Fig. 13k). In the same year, Qin-Teng Lai et al.141 prepared pressure sensors with embossed microstructures using the pencil graphite friction (PGF) method (Fig. 13l). This method is simple, costeffective, and environmentally friendly. The resulting graphite paper-based sensors exhibited a maximum sensitivity of 7202.2 kPa⁻¹ (Fig. 13m) and were capable of real-time detection of human physiological signals such as wrist pulse and finger joint flexion, providing valuable information for disease prevention and diagnosis.

Paper-based resistive pressure sensors offer significant advantages in terms of low cost, flexibility, wearability, and environmental friendliness. However, they face challenges in terms of environmental adaptability. Additionally, improving immunity to interference is an area that requires further development. While current research has focused on enhancing sensitivity and stability, minimizing the impact of external environmental factors on sensor performance remains a crucial topic 142,143. For example, improving moisture and corrosion resistance while maintaining sensitivity is a key direction for future research.

Paper-based capacitive stress sensors. Compared with resistive sensors, capacitive sensors demonstrate significant advantages in multiple aspects, such as high impedance, low power consumption, good temperature stability, simple structure, strong adaptability, good dynamic response, and the ability for non-contact measurement. These advantages make capacitive sensors widely applicable in various fields and hold great promise for future applications. For example, Pengfei

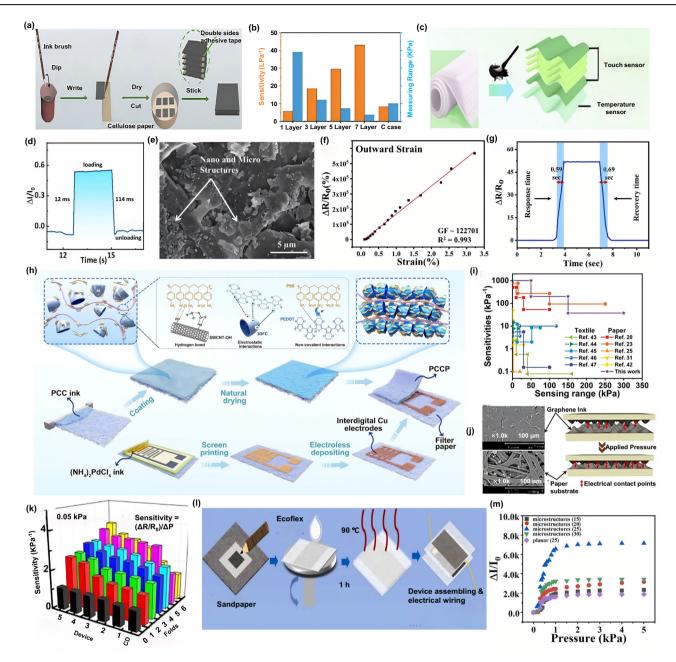


Fig. 13 | Paper-based resistive stress sensors. a Fabrication flow chart of graphene-paper-based pressure sensor. b Sensitivity and response range¹³⁷. c Schematic illustration, photos for the fabrication of the touch-temperature sensor. d Based on kirigami art and landscape ink painting for touch sensors²³⁴. e SEM micrographs of graphite coated cellulose paper frameworks. f Response of sensor upon provision of bending strain up to 3.23% for inward. g Graph showing the response and recovery time of the demonstrated sensor¹³⁸. h Schematic illustration of

fabricating an all-paper pressure sensor by integrating a blade-coated top sensing electrode and a screen-printed bottom copper electrode. i Sensitivity and sensing range of all-paper pressure sensor compared to other paper-based and textile-based sensors reported in literatures 139 . j ECR variation mechanism for tactile sensing. k Sensitivity data of scaled down devices 140 . 1 Schematic diagram of the fabrication process of graphite-based e-skin developed by PGF approach. m Sensitivity of five different e-skins 141 .

Zhao et al.¹⁴⁴ realized for the first time an all-paper shape designable capacitive pressure sensor by using multilayer thin paper as dielectric and flexible polypyrrole (PPy) printed paper as electrodes (Fig. 14a). This innovative design not only reduces cost but also provides unique features such as stretchability and 3D sensing. It outputs a stable capacitive response under dynamic pressure (Fig. 14b), demonstrating excellent pressure sensing performance and proximity sensing capability. Hua Xue et al.¹⁴⁵ constructed a capacitive pressure sensor with a double electric layer (EDL) structure by introducing ionic liquids (ILs) (Fig. 14c). This sensor achieves ultra-high sensitivity (Fig. 14d) and an ultra-low detection limit of 2 Pa (Fig. 14e). Azmal Huda Chowdhury et al.¹⁴⁶ further

simplified the sensor fabrication process by preparing a thin, paper-based capacitive pressure sensor using textiles (Fig. 14f), which is easy and inexpensive to produce, and achieving a high sensitivity of 2.9 kPa⁻¹ over the pressure range of 0–16 kPa and a wide detection range (Fig. 14g), which can effectively measure pulse waveforms. Myda Arif et al. ¹⁴⁷ developed a capacitive sensor-based mouse pad using a sensing array on paper (Fig. 14h). This design allows for mouse operations including up, down, left, right, and left and right buttons. Additionally, it addresses the issue of high pressure required by resistive mouse pads while providing fast response, high durability, and excellent optical performance. In order to further reduce the manufacturing cost of the sensor and simplify the

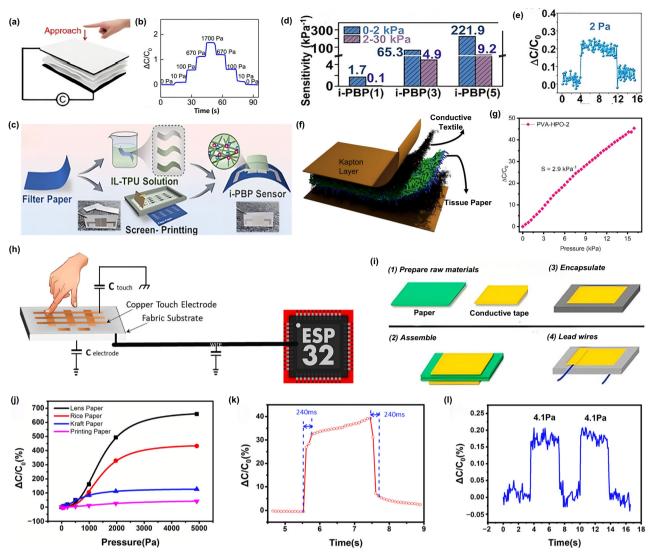


Fig. 14 | Paper-based capacitive stress sensors. a Schematic diagram of capacitive sensors. b Reversible and subtle pressure responses 144 . c Schematic diagram of the fabrication process and optical photos of the i-PBP sensors. d Sensor in different pressure ranges of sensitivities. e Response-recovery curve of i-PBP sensor under pressure of 2 Pa 145 . f Schematic of the tissue-paper-based supercapacitive pressure sensor. g Detailed relative capacitance changes over pressure for the sensor with the

highest electrolyte concentration¹⁴⁶. **h** Side view representation of a flexible hybrid design capacitive sensor¹⁴⁷. **i** Fabrication process of capacitive pressure paper-based sensors. **j** Comparison of sensitivity curves of capacitive pressure sensors made from four types of paper materials. **k** Response time curve of the rice paper-based capacitive pressure sensor. **l** Minimum pressure resolution curve of the rice paper-based capacitive pressure sensor⁵⁴.

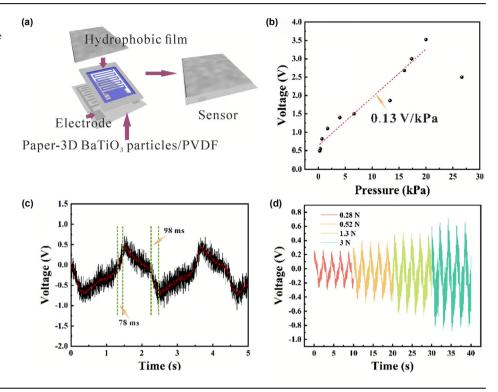
preparation process, Haozhe Zhang et al.⁵⁴ utilized the inherent micronano structure of paper to prepare a capacitive paper-based pressure sensor, using paper as both a flexible substrate and a sensitive element (Fig. 14i). This sensor achieved a high sensitivity of 1.623 kPa⁻¹ (Fig. 14j), a fast response time of 240 ms (Fig. 14k), and a minimum pressure resolution of 4.1 Pa (Fig. 14l). It also maintained a stable capacitive output over multiple load cycles, enable accurate and stable real-time monitoring of human wrist movements.

Paper-based capacitive stress sensors offer advantages such as high accuracy, good stability, fast dynamic response, low power consumption, and easy integration. However, they also have disadvantages, including high output impedance, poor loading capability, parasitic capacitance effects, and stringent requirements for paper-based materials¹³². Therefore, when selecting and using paper-based capacitive pressure sensors, these factors need to be considered comprehensively and weighed against specific application scenarios.

Paper-based piezoelectric stress sensors. Paper-based piezoelectric stress sensors are attracting attention in the field of flexible electronics

due to their unique properties. These sensors utilize the principle of charge generation by piezoelectric materials to convert mechanical stress into electrical signals 148,149. They have gained the attention of researchers due to their high sensitivity, fast response time, and wide range of applications in microstress detection and human motion monitoring. In recent years, several studies have focused on developing paper-based piezoelectric sensors. In 2023, Chenxu Yu et al. 150 constructed a paperbased piezoelectric sensor with an irregularly porous structure using a polycarbonate (PC) film as the outer protective layer of a BPP-PC sensor (Fig. 15a), which was capable of sensing an external micro-pressure of at least 0.1 mg and has a sensitivity of 0.13 V/kPa, a pressure range of 0-20 kPa, and a response time of 78 ms (Fig. 15b, c). Additionally, it demonstrated excellent detection of water droplets and sensitively captured human motions such as footsteps, hand clapping, wrist and finger bending, showing potential for detecting minute pressures and monitoring human motions (Fig. 15d). However, the piezoelectric coefficients of organic piezoelectric materials and piezoelectric composites are significantly lower than those of conventional piezoelectric ceramics, which limits the sensitivity of pressure sensor arrays¹⁵¹. Furthermore, despite the

Fig. 15 | Paper-based piezoelectric stress sensors. BPP-PC sensor picture (a), sensitivity (b), response time (c), output voltage (d) at different low voltages. ¹⁵⁰.



superior electromechanical properties of piezoelectric ceramics such as lead zirconate titanate (PZT), their inherent stiffness and brittleness restrict their application in flexible electronics¹⁵². Therefore, although paper-based piezoelectric pressure sensors hold great potential in flexible electronics, further research is needed to overcome these challenges and enable wider applications.

Paper-based mechanical sensors, a new type of sensor with unique advantages, show broad application prospects in wearable devices, health monitoring, and environmental monitoring ^{153,154}. However, different types of paper-based mechanical sensors have their own limitations. Moreover, gases, humidity, and strain often coexist and interact with each other. Since paper itself is hydrophilic and flexible, developing high-performance paper-based sensors remains challenging. Although paper-based mechanical sensors have great potential for application in various fields, continuous improvement and optimization of material selection, structural design, and manufacturing processes are needed to better meet real-world demands.

Paper-based gas sensor

Gas sensors are widely applied in various fields such as industry, homes, and environmental monitoring because they can detect the types and concentrations of gases in the environment. To reduce health risks and protect the environment, it is necessary to develop gas sensors that are highly responsive, sensitive, selective, and stable, while also maintaining low cost and low power consumption¹⁵⁵. Paper-based gas sensors typically use active materials (such as metal oxides, carbon-based materials, and composite materials) as sensing elements. When gas molecules come into contact with these active materials, chemical or physical reactions occur, leading to changes in the materials' resistance, conductivity, or other properties¹⁵⁶. In recent years, paper-based gas sensors have been extensively studied in many fields. For example, Ronil J. Rath et al. 157 prepared a paper-based gas sensor by coating polystyrene sulfonate (PSS) on paper to detect and distinguish between ammonia and carbon dioxide gases (Fig. 16a). Due to the interaction between PSS and gas molecules, when NH3 or CO2 comes into contact with PSS in a humid environment, chemical reactions occur, leading to changes in the sensor's resistance. The sensor exhibited significant resistance changes to ammonia and carbon dioxide under high humidity conditions (>85%), achieving high selectivity detection and a wide detection

range (ammonia 0.25 ppm to 2500 ppm) in complex environments, providing an important foundation for gas detection in practical applications (Fig. 16b). Xiao Ye et al. 158 prepared a fully inkjet-printed chemiresistive sensor array by coating molecularly imprinted sol-gel (MISG) active material and Ketjen black (KB) ink on paper (Fig. 16c) to detect and distinguish volatile organic acids (VOAs). Due to the molecular imprinting effect of the HA-MISG layer for recognizing target HA vapor, the expansion effect caused by recognition increases the distance between conductive KB particles, leading to an increase in resistance. When the HA-MISG/KB sensor is cleaned, air removes the target molecules from the imprinted cavities of the HA-MISG layer, thus reducing the resistance. The sensor achieved highly cross-selective detection of VOAs at room temperature with high sensitivity and selectivity, as well as low detection limits (Fig. 16d). Additionally, the proposed sensor array demonstrated strong sensor robustness, with excellent stability, flexibility, and humidity resistance. Guh-Hwan Lim et al.¹⁵⁹ developed a paper-based sensor for self-activated chemiresistive gas detection by fabricating boron nitride/carbon nanotube (BNCNT) composite paper to detect nitrogen dioxide (NO₂) gas (Fig. 16e). The sensor achieves highly sensitive detection of gases through the selfheating effect and changes in electrical conductivity of the boron nitride/ carbon nanotube composite paper. The sensor exhibited high sensitivity, selectivity, and fully reversible response characteristics, achieving highly selective detection of NO2 gas at room temperature with a detection limit as low as 3.41 ppb. Moreover, the sensor showed excellent long-term stability and environmental adaptability, providing a promising platform for environmental monitoring in smart clothing and wearable electronic devices

Paper-based gas sensors have shown promising applications in several fields due to their low cost, biodegradability and high sensitivity. However, despite their significant advantages in providing fast response and high sensitivity, these sensors still face challenges in terms of stability and environmental adaptability. For example, paper-based sensors have poor durability, are susceptible to humidity variations, and can fail in high humidity or underwater environments¹⁶⁰. Therefore, future research needs to optimize material selection and sensor design and explore new materials and technologies to enhance the performance and utility of paper-based humidity sensors.

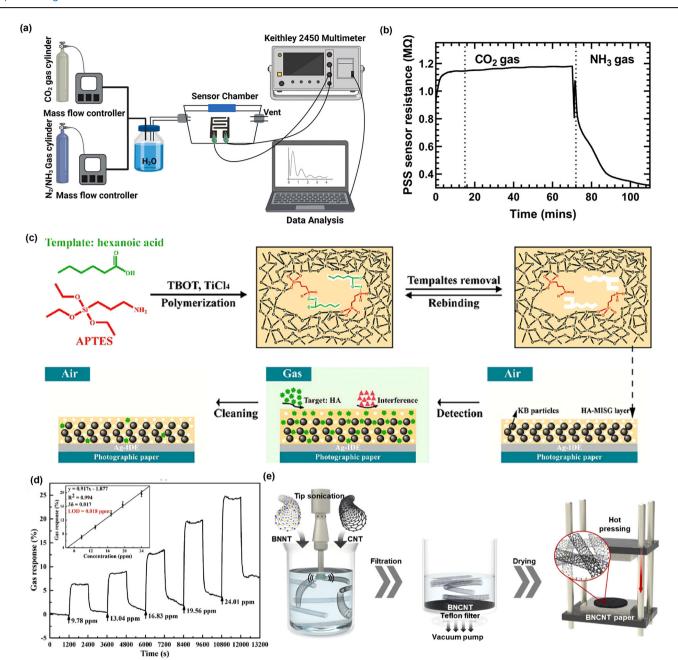


Fig. 16 | Paper-based gas sensor. a Schematic set-up for testing PSS sensor performance in the gas phase. **b** PSS sensor response to humidified nitrogen, carbon dioxide, and ammonia gas¹⁵⁷. **c** HA-MISG/KB sensors for selective HA vapor

detection. **d** Response curve of HA-MISG/KB sensor to different concentrations of Hexanoic Acid (HA)¹⁵⁸. **e** Schematic illustration for the fabrication process of the BNCNT hybrid papers¹⁵⁹.

Paper-based humidity sensors

Humidity is a key factor in many fields including ecology, industry, medicine and indoor/outdoor environmental monitoring. Additionally, high humidity levels can make breathing difficult and cause coughing and wheezing in asthmatics. High levels of humidity can create a favorable environment for dust mites and mold to thrive in the home, which can be harmful to human health¹⁶¹. Due to their low cost, biodegradability, and environmental compatibility, paper-based humidity sensors have become a hot research topic in flexible electronic devices. Recent research advances have shown that paper-based humidity sensors have significantly improved in terms of response speed, sensitivity, and stability. For example, Penghui Zhu et al.¹⁶² prepared a paper-based bilayer humidity sensor (PBHS) (Fig. 17a) with high sensitivity and good durability. The large number of hydrophilic hydroxyl groups on the surface of CNF and paper fibers allows rapid exchange of water molecules between the moisture-sensitive material

and the external environment through hydrogen bonding, the prepared sensor has a 95% relative humidity with a 65.0% maximum response value (Fig. 17b), and also exhibited a bendability with a maximum curvature of 22.2 cm⁻¹ and up to 50-fold folding durability (Fig. 17c). Zhi Song et al. ¹⁶³ developed a paper-based capacitive humidity sensor based on graphene oxides (Fig. 17d). This sensor achieves ultra-high sensitivity (Fig. 17e) with an optimized structural design and fast response/recovery times of 170/40 s (Fig. 17f). It is suitable for respiratory monitoring, non-contact applications, and food safety monitoring. Xiaoqiang Li et al. ¹⁶⁴ prepared a self-powered humidity sensor using drawing carbon ink (CI), cellulose filter paper (FP), and polyester conductive tape (Fig. 17g). This sensor generates a humidity-dependent voltage by adsorbing water molecules in a humid environment. It exhibits excellent humidity-voltage responsiveness and cycling performance over a wide humidity monitoring range (11–98%) (Fig. 17h), making it suitable for monitoring human health, air humidity, and non-contact

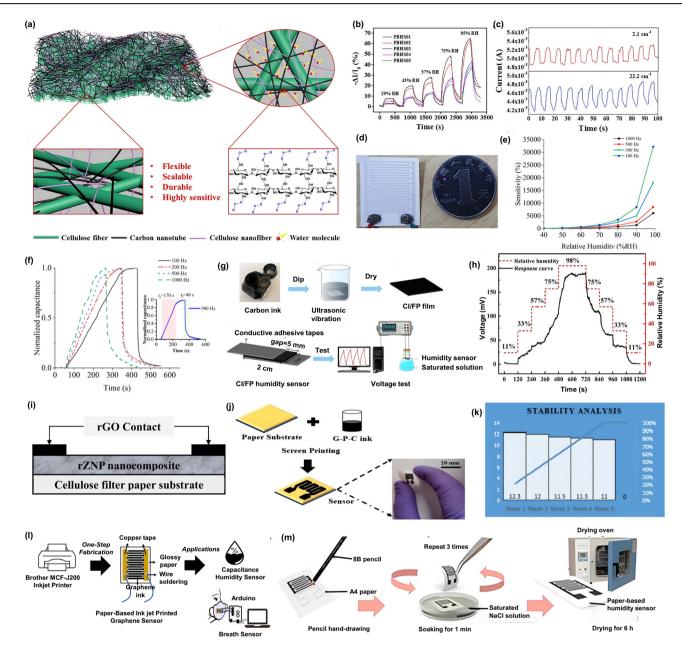


Fig. 17 | Paper-based humidity sensors and the device performance. a Schematics illustrating the mechanism of CNF/CNT dual network-enabled flexible humidity sensors with high sensitivity and good durability. **b** Dynamic response and recovery curves of the PBHS with various CNF/CNT loadings between 11 and 29, 43, 57, 75, and 95% RH. **c** Current-time curves of human nose breathing under bending states with curvatures of 2.1 and 22.2 cm⁻¹, respectively¹⁶². **d** Fabricated sensor. **e** Sensitivity at different operation frequencies. **f** Response/ recovery at different

operation frequencies for the GO sensor with concentrations of 1 mg/mL¹⁶³. **g** Fabrication process of self-powered CI/FP humidity sensor. **h** The sensor dynamic voltage response curve increases with RH from 11% to 98% and then decreases to 11%¹⁶⁴. **i** Cross-sectional image based on rZNP humidity sensor¹⁶⁵. **j** Screen-printed G-P-C humidity sensor. **k** Stability analysis plot¹⁶⁶. **1** Sensor schematic diagram of graphene ink inkjet printing paper¹⁶⁷. **m** Manufacturing process of pen and paper hydration sensor¹⁶⁸.

humidity sensing. A.S.R.A. Subki et al.¹⁶⁵ prepared resistive humidity sensors based on rGO/ZNP nanocomposites (rZNP) with different rGO loadings (Fig. 17i). The presence of oxygen vacancy defects and oxygen-related chemical bonding on the surface of rZNP enhances the sensor's sensitivity to water molecules, resulting in higher sensitivity compared to conventional humidity sensors. These sensors also demonstrate excellent repeatability and good stability.

Disposable sensors play a crucial role in monitoring respiratory and ambient humidity. Parthasarathy P et al. ¹⁶⁶ fabricated cost-effective and flexible disposable humidity sensors using biocompatible and conductive graphene-polypyrrole-carbon black (G-P-C) inks on paper substrates (Fig. 17j). These sensors exhibit high flexibility,

stability, and repeatability (Fig. 17k), and can be used to monitor soil moisture and space humidity. In addition, Lim W Y et al. ¹⁶⁷ designed an inexpensive, disposable, and sensitive capacitive humidity sensor using a fork-finger electrode (IDE) configuration for respiratory sensing (Fig. 17l). This sensor can continuously and real-time monitor respiration in three different scenarios (e.g., normal breathing, deep breathing, and coughing) and exhibits a short response and recovery time of less than 5 s. Guangyu Niu et al. ¹⁶⁸ prepared a highly sensitive disposable paper-based humidity sensor by treating a hand-drawn graphite electrode on cellulose paper with a saturated NaCl solution (Fig. 17m). The resultant sensor features high sensitivity, and a wide relative humidity (RH) range from 5.6% to 90%, enabling convenient

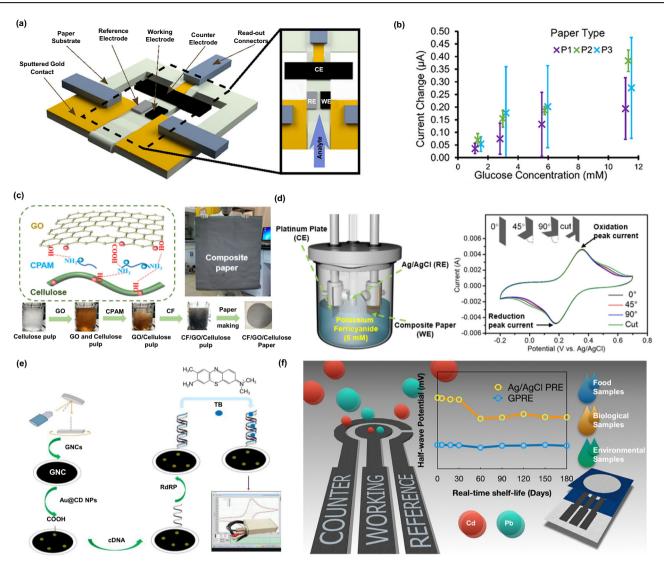


Fig. 18 | Paper-based current-based electrochemical sensors. a Overall architecture of the electrochemical sensors developed in this work. b Change in current as a function of glucose concentration ¹⁸³. c Fabrication of CF/GO/cellulose paper. d CV

of potassium ferricyanide 170 . e Schematic diagram of GNCs/ paper electrochemical gene sensor electrodes 184 . f Comparison of electrochemical properties and stability of GPRE and conventional Ag/AgCl PRE 185 .

monitoring of respiratory rate and non-contact finger position. Moreover, the preparation method is simple and low-cost.

Paper-based electrochemical sensors

Paper-based electrochemical sensors utilize a paper substrate as an electrochemical sensing platform. These sensors are inexpensive, portable, user-friendly, and are suitable for implementation in resource-limited environments, such as emerging economies or field environments 169,170. Electrochemical sensors typically consist of three electrodes: a working electrode, a counter electrode, and a reference electrode. The working electrode, where the reaction occurs, can be modified to enhance recognition and sensitivity to specific substances. For example, immobilizing functional groups or biorecognition elements (e.g. antibodies or enzymes) on the working electrode enables the electrochemical detection of specific species 171,172. The working principle is based on the redox reaction of the target substance on the electrode surface, generating electrochemical signals such as current, potential, or charge. These signals are closely related to the concentration or nature of the target substance, and by detecting changes in these signals, qualitative and quantitative analyses can be achieved 173,174. In the early stage, Dungchai et al. 175 described the paper electrochemical analysis device. Since then, the paper electrochemical analysis device has been widely popularized due to its advantages such as simple, low power consumption, low detection limit and easy quantification. Compared to conventional high-pressure liquid chromatography and gas chromatography coupled with mass spectrometry, paper-based electrochemical sensors achieve high analytical sensitivity due to the many nanoparticles that can increase the loading capacity and the wide surface area of reactant mass transfer¹⁷⁶. Wearable paper-based electrochemical sensors have potential applications in monitoring temperature 177, pressure 178, humidity and respiration 179, and healthcare (e.g., monitoring specific analytes, diabetes, or cardiovascular diseases)¹⁸⁰. Paper-based electrochemical biosensors can be categorized into three main types based on their operating principles: amperometric (current-based), potentiometric (potential-based), and impedimetric (resistance-based). Each type has its own characteristics and is suitable for different detection needs and application scenarios. Amperometric sensors focus on current changes, potentiometric sensors on potential changes, and impedimetric sensors on resistance changes. These differences allow for the selection of the most suitable sensor type for specific applications. These sensors can be used in clinical diagnosis, environmental monitoring, and food safety¹⁸¹.

Paper-Based Current-Based Electrochemical Sensors. The operating principle of paper-based current-based electrochemical sensors

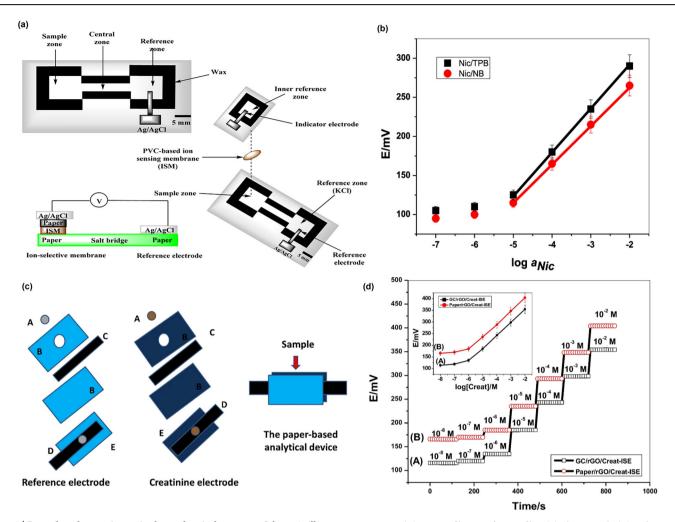


Fig. 19 | Paper-based potentiometric electrochemical sensors. a Schematic illustrations of a referenced electrochemical paper-based electrochemical device.

b Potentiometric response behavior of the presented nicotine-sensing electrochemical paper-based device¹⁸⁷. c Schematic diagram of creatinine potentiometric

paper sensor :(A) sensing film or reference film, (B) plastic mask, (C) carbon nanotube conductive paper, (D) electrical contact point, (E) final sensor view. ${\bf d}$ Calibration diagram of the proposed creatinine paper sensor: (A) GC/rGO-Creat-ISE; (B) paper/rGO-Creat-ISE.

primarily relies on electrochemical reactions. When the measured substance reacts with the sensitive electrode inside the sensor, a change in current is generated, which is directly related to the concentration or nature of the measured substance. By measuring this change in current, quantitative or qualitative analysis of the measured substance can be achieved¹⁸². In recent years, paper-based current-based electrochemical sensors have demonstrated significant scientific value and application potential in the field of electrochemical detection and analysis due to their low cost, simple preparation process, high scalability, and good sustainability. For example, Christopher J. Valentine et al. 183 used laser cutting to define the electrode region, cleverly fabricating a paper-based electrochemical sensor (Fig. 18a). The sensor leverages the porosity of paper to provide microchannels that respond differently to varying glucose concentrations. Changing the paper's porosity results in nearly a 2-fold increase in current change (Fig. 18b). Cheng Wang et al. 170 prepared functional composite paper for electrochemical detection using traditional papermaking methods (Fig. 18c). This composite paper exhibits good controllability, stability, and reproducibility. It shows similar cyclic voltammetry (CV) curves for electrodes that are folded or even cut at different angles (Fig. 18d). The sensor demonstrates remarkable sensitivity, capable of accurately detecting phenolic compounds at low concentrations. Specifically, the detection limits are as low as 0.045 mg/L for hydroquinone (HQ), 0.093 mg/L for p-chlorophenol (CP), and 0.571 mg/L for p-nitrophenol (NP). These low detection limits highlight the sensor's significant potential for trace analysis applications. Mohammad Ali Farzin et al. 184 developed a highly sensitive and selective electrochemical current-based sensor using conductive graphite nanocrystals (GNC) (Fig. 18e). The sensor demonstrates high stability, low limit of detection (LOD), and high recoveries of spiked sputum samples for the determination of the SARS-CoV-2 RdRP gene. The use of GNC effectively improves the paper surface and increases electrical conductivity. Prasongporn Ruengpirasiri et al. 185 integrated a graphene pseudo-reference electrode (GPRE) with a paper-based electrochemical device (PED) to create an electrochemical sensor for the sensitive and selective detection of heavy metals cadmium (Cd) and lead (Pb) (Fig. 18f). The sensor demonstrates remarkable analytical performance. For cadmium ions (Cd²⁺) and lead ions (Pb²⁺), the detection limits are as low as 0.69 ng/mL and 5.77 ng/mL, respectively, with electrochemical of 70.16 $\mu A \, mL \, \mu g^{-1} \, cm^{-2}$ Cd^{2+} sensitivities for 38.34 μ A·mL μ g⁻¹ cm⁻² for Pb²⁺.

Paper-based potentiometric electrochemical sensors. Paper-based potentiometric electrochemical sensors have gained widespread attention in recent years due to their low cost, portability, biodegradability, environmental friendliness, and ease of fabrication. These sensors are particularly easy to handle and highly active, making them well-suited for point-of-care and field applications. The operating principle of these sensors is based on electrochemical reactions and potential

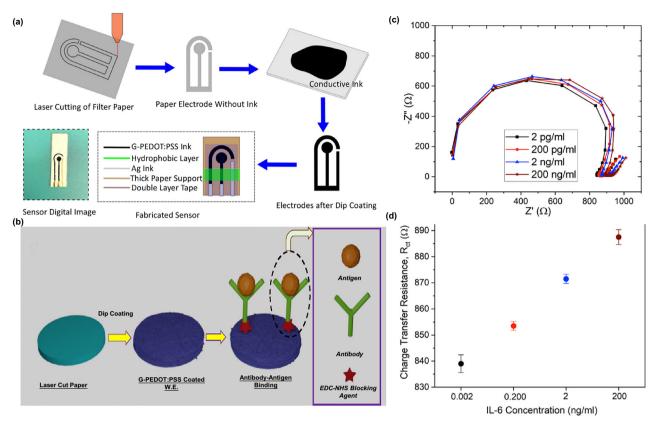


Fig. 20 | Paper-based resistive electrochemical sensors. a Paper-based biosensor fabrication. b Schematic representation of TNF-α/IL-6 detection protocol for paper-based electrodes. c EIS characterization of IL-6 detection in human serum from

2 pg/mL to 200 ng/mL. $\bf d$ Charge transfer resistance with uncertainty plot for IL-6 concentration from 2 pg/mL to 200 ng/mL 189 .

measurements. When a substance reacts with a sensitive electrode within the sensor, voltage changes are generated that are directly related to the concentration or nature of the substance. By measuring these potential changes, quantitative or qualitative analysis of the measured substance can be achieved¹⁸⁶. In recent years, several studies have been conducted on the development of paper-based potentiometric electrochemical sensors. For example, Abd El-Galil E. Amr et al. 187 developed a paperbased nicotine sensor with an integrated miniaturized paper reference electrode (Fig. 19a). Compared to conventional electrochemical sensors, this sensor does not require storage of reference electrodes and utilizes ionophores combined with conductive polymers for potentiometric measurements. It exhibits good performance over a wide linear range of 6.0 µm-8.0 µm, and a stable potentiometric response for different nicotine concentrations (Fig. 19b). The sensor exhibits high responsiveness to nicotine concentration changes, with sensitivities of 55.2 ± 0.3 mV/decade for the [Nic/TPB] system and 51.2 ± 0.6 mV/decade for the [Nic/NB] system. Ayman H. Kamel et al. 188 prepared a novel paper-based potentiometric electrochemical sensor by combining a reference Ag/AgCl electrode with a creatinine-selective electrode (Fig. 19c). This sensor demonstrates high analytical throughput, excellent accuracy, superior repeatability, stable response, and good selectivity for direct potentiometric detection of creatinine in various human urine samples. It shows a sensitivity of $56.5 \pm 1.1 \text{ mV/decade}$ ($R^2 = 0.9998$) with a detection limit of 1.0 µm (Fig. 19d).

Paper-based resistive electrochemical sensors. Paper-based resistive electrochemical sensor is an electrochemical sensor that combines paper-based material with a resistive detection principle. Its working principle is based on the fact that when the substance to be measured undergoes an electrochemical reaction on the electrode surface, it causes

a change in the resistance between the electrodes, which can be detected. When the substance to be measured interacts with the active sites on the electrode surface, it changes the electronic state and charge distribution on the electrode surface, resulting in a change in the resistance between the electrodes. By measuring this resistance change, the concentration or nature of the substance to be measured can be indirectly determined ^{172,173}. In 2023, Md Ashiqur Rahman et al. 189 fabricated miniature paper-based biosensors (GCPPS) (Fig. 20a) with graphene-PEDOT:PSS coating for the detection of dopamine, TNF-α and IL-6 (Fig. 20b). Their study showed that the charge transfer resistance increased with the concentration of IL-6 (Fig. 20c). Moreover, a linear relationship was observed between the charge transfer resistance and IL-6 concentration in human serum (Fig. 20d). The sensor exhibits excellent sensitivity and is capable of detecting a variety of biomolecules. For dopamine, the detection range is 12.5-400 µM with a detection limit (LOD) as low as 3.4 µM. For tumor necrosis factor- α (TNF- α), the detection range is 0.005-50 ng/mL with an LOD of 5.97 pg/mL. For interleukin-6 (IL-6), the detection range is 2 pg/ $mL^{-2} \mu g/mL$ with an LOD of 9.55 pg/mL.

In summary, paper-based electrochemical sensors are a novel type of sensor that offer low cost, high sensitivity, and portability. These attributes endow them with broad application prospects in the fields of biomedicine, environmental monitoring, and food safety, as well as in point-of-care testing (POCT). By integrating with other electronic components and adopting miniaturized designs, portable and intelligent detection systems can be developed, providing more convenient and efficient detection methods for biomedicine, environmental monitoring, and food safety. However, despite their many advantages, paper-based electrochemical sensors still face some challenges in practical applications. With the continuous advancement of materials science, nanotechnology, and electrochemical sensing techniques, it is anticipated that paper-based

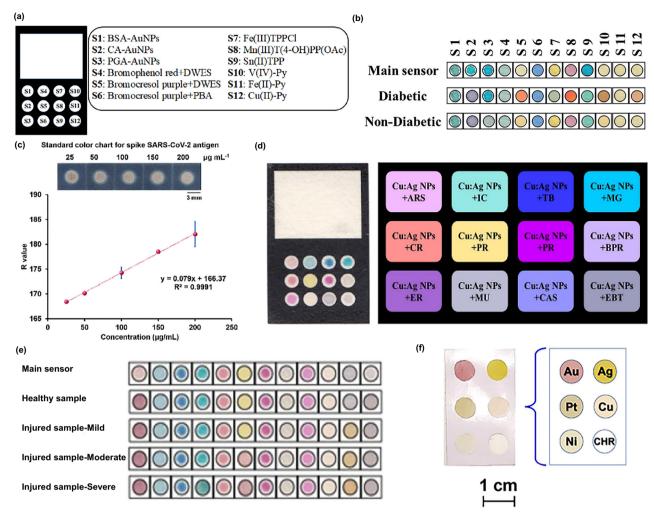


Fig. 21 | Paper-based colorimetric sensors. a Proposed mode of sensor and list of sensing elements¹⁹⁶. **b** Color response of the sensor after exposure to saliva metabolites and color map extracted by image analysis¹⁹⁷. **c** Relationship between colorimetric sensors and SARS-CoV-2 antigen concentration standard color chart¹⁹⁸.

d Sensor-based template and color receptor list. **e** Colorimetric responses and colorimetric difference maps of plasma sample metabolites associated with the sensor for healthy participants and injured persons with varying injury severity¹⁹⁹. **f** Color response of the sensor array exposed with different concentrations of PAA²⁰⁰.

electrochemical sensors will achieve higher sensitivity, a wider measurement range, and greater durability in the future.

Paper-based optical sensors

Paper-based optical sensors are a type of optical sensor that utilize paper or a paper substrate as their foundation. These sensors leverage the unique characteristics of paper in combination with optical principles to achieve sensing functions. The working principle primarily relies on optical phenomena, such as the absorption, reflection, and scattering of light. When light strikes the sensor, the material on the sensor's surface interacts with the light to generate specific optical signals. These signals can then be converted into electrical or other forms of signals for subsequent measurement and analysis ^{190,191}. Generally, paper-based optical sensors are categorized into two main types: paper-based colorimetric sensors and paper-based fluor-escent sensors.

Paper-based colorimetric sensors. Paper-based colorimetric sensors are a type of optical sensor that detects color changes in substances and quantitatively analyzes the concentration of target substances. They primarily operate based on color changes caused by chemical reactions. When a sample to be detected comes into contact with a chemical reagent on the paper-based material, a specific chemical reaction occurs, resulting in a color change. This color change can be quantitatively analyzed by colorimetry or even observed with the naked eye

to determine the concentration of the target substance¹⁹². Paper-based colorimetric sensors have a wide range of applications, including but not limited to the absorption and concentration measurement of food dyes¹⁹³, detection of hydroperoxide-related biomarkers¹⁹⁴, and monitoring biogenic amines in food spoilage¹⁹⁵. In recent years, several studies have been conducted on the development of paper-based colorimetric sensors. For example, Mohammad Mahdi Bordbar et al. 196 prepared a paper-based colorimetric sensor (Fig. 21a) for detecting COVID-19 using nanoparticles, organic dyes, and metal-ion complexes as sensing elements to capture metabolites in human serum samples. The sensor exhibits a sensitivity of 83.9% for detecting healthy individuals and 82.2% for detecting those infected with COVID-19. It can display distinct colors for infected and control samples and estimate the severity of viral infections. Hosseini M S et al. 197 developed a colorimetric sensor based on a folded paper structure to monitor metabolic changes due to diabetes. Using porphyrins, pH-sensitive dyes, and silver nanoparticles, the sensor could show different colors in response to the chemical composition of saliva samples from diabetic and non-diabetic individuals (Fig. 21b). Pornchanok Punnoy et al. 198 prepared a paper-based colorimetric sensor using gold nanoparticles (AuNPs) for rapid visual detection and sensitive quantitative detection of the SARS-CoV-2 antigen (Fig. 21c). The sensor demonstrates remarkable sensitivity for the detection of SARS-CoV-2 antigen and urea. For SARS-CoV-2 antigen, the linear range is from $25\,\mu\text{g/mL}$ to

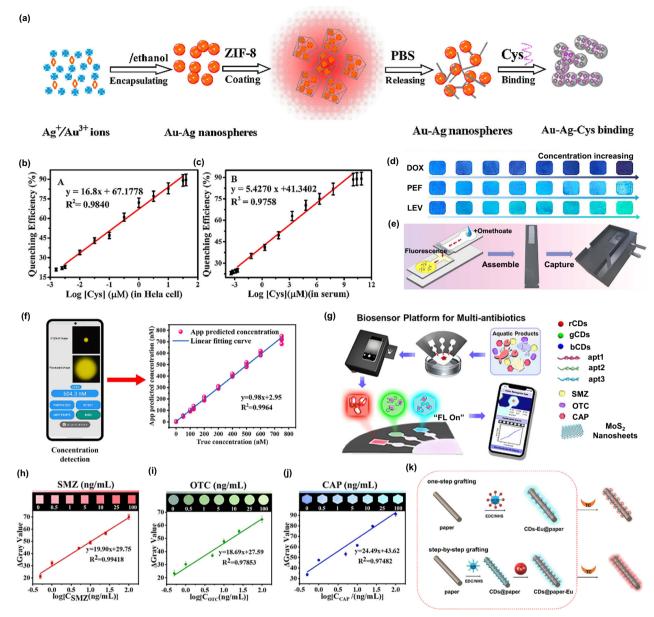


Fig. 22 | Paper-based fluorescent sensors. a Schematic diagram of main procedures and mechanisms of Cys detection using fluorescent Au-Ag@ZIF-8 test paper. b, c Quenching efficiency and calibration curves of logarithmic concentrations of targeted Cys added to cell extraction (b) and human serum sample (c)²⁰⁹. d Numerical signals output by the sensor at different DOX/PEF/LEV concentrations²¹⁰. e, f Construction of a microfluidic paper fluorescence sensor based on a smartphone sensing platform and an application diagram for predicting the

concentration of omethoate¹⁹¹. **g** The mCD- μ PAD aptamer sensor and portable detector simultaneously visually detect multiple antibiotics in devices with smartphones. **h**-**j** Fluorescence images of the sensor and calibration curves between Δ gray values and logarithmic SMZ, OTC, and CAP concentrations in the red, green, and blue detection regions²¹⁰. **k** Schematic diagram of preparation process of CDs-Eu@paper and CDs@paper-Eu²¹¹.

200 µg/mL, with a detection limit of 13.75 µg/mL and a quantification limit of 45 µg/mL. Mohammad Mahdi Bordbar et al. 199 immobilized copper-silver bimetallic nanoparticles (BMNPs) containing organic dyes and coated with dobutamine on paper to create a colorimetric sensor (Fig. 21d). The sensor demonstrates remarkable sensitivity, with a linear range from 25 µg/mL to 200 µg/mL, a detection limit of 13.75 µg/mL, and a quantification limit of 45 µg/mL. Additionally, the sensor's quantitative response shows a strong correlation with the severity of the injury (Fig. 21). The sensor's response can be observed with the naked eye within 3 min, and it achieves an accuracy of 87% in distinguishing between injured and healthy individuals. Annalisa Scroccarello et al. 200 used laser-induced generation of different types of metal nanoparticles (LIMs) on paper to prepare paper-based colorimetric sensor arrays (Fig. 21f) for detecting peroxyacetic acid (PAA)

vapor. These sensor arrays could discriminate PAA exposure levels from 0.5 to 50 mg/m 3 and enabled fast organic dye-catalyzed conversion and sensitive PAA detection (LOD \leq 0.3 μ g/mL).

Paper-based fluorescent sensors. The paper-based fluorescence sensor is a paper-based optical sensor whose working principle is mainly based on the interaction between the fluorescent nanomaterial and the object to be measured. When the excitation light irradiates the fluorescent nanomaterials, the electrons in the material absorb energy and jump to an excited state. Subsequently, part of the energy is released as the electrons return to the ground state, generating fluorescence in the process. When the analyte reacts with the fluorescent nanomaterials, the fluorescence properties (e.g., fluorescence intensity, wavelength, etc.) of the fluorescent nanomaterials change, thereby enabling the detection of the target

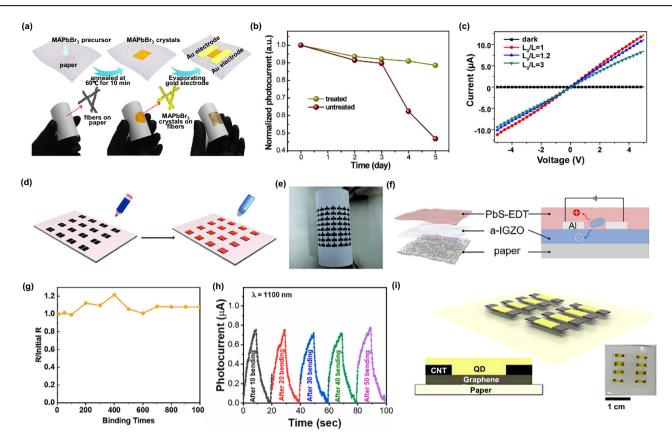


Fig. 23 | **Paper-based photodetectors.** a Schematic diagram of the preparation process of the PBPPD. **b** Photocurrent changes of both the treated and untreated devices being placed in an environment of 60% relative humidity for 120 h. c I-V curves of the PBPPD at different bending angles under the illumination of 2.06 μW input power²¹². **d** Schematic illustration of the pen-writing photodetectors. **e** Photograph of the pen-written perovskite photodetector arrays²¹³. **f** The schematic

diagram of the a-IGZO/PbS QDs-EDT heterojunction deposited on the Senyan paper. ${\bf g}$ Change curves of R/Initial R with bending times of the Senyan paper-based device 214 . ${\bf h}$ Photoresponse test for flexibility and durability of WS $_2$ /Ti $_3$ C $_2$ T $_x$ heterogeneous functionalized paper-based photodetectors 215 . ${\bf i}$ Schematics and images based on QD/SLG PD on a paper substrate 216 .

analyte²⁰¹⁻²⁰⁵. With the advantages of high sensitivity, fast response, and multiple detection, paper-based fluorescent sensors hold broad application prospects and development potential in the fields of biomedicine, environmental monitoring, and food safety²⁰⁶⁻²⁰⁸. For example, Yuanyuan Cai et al.²⁰⁹ developed a fluorescence-based sensing test paper (Fig. 22a) using gold and silver nanospheres and ZIF-8 MOF for rapid, simple, and sensitive detection of cysteine (Cys) in HeLa cells (as low as ~2.0 nM) and Cys levels in serum (Fig. 22b, c), with high analytical selectivity, sensitivity and reproducibility, making it suitable for rapid on-site monitoring of Cys in clinical laboratories. Tingting Li et al.²¹⁰ prepared a paper-based fluorescent sensor (Fig. 22d) using multi-doped graphene quantum dots (M-GQD) for the highly sensitive detection and discrimination of tetracyclines (TCs), aminofluoroquinolones (AFQ), and ofloxacin-like quinolones (OFQs). This sensor showed different responses to the three antibiotics, with limits of detection (LOD) of 39.30 nM, 48.60 nM, and 35.10 nM, respectively, indicating excellent detection limits for antibiotics. Shuai Liu et al.¹⁹¹ prepared a microfluidic paper-based fluorescent sensor by combining fluorescent microfluidic paper strips with a smartphone-based sensing platform (Fig. 22e) for rapid and sensitive detection of omethoate pesticides. The sensor achieved a high regression coefficient of 0.9964 over the omethoate concentration range of 0-750 nM (Fig. 22f) and enabled rapid on-site detection of finished pesticides in real samples in less than 10 min, with results comparable to those obtained by standard methods.

Paper-based fluorescent sensors have also demonstrated broad applications and significant development potential in aquaculture. Xinyu Tong et al.²¹⁰ designed a paper-based fluorescent sensor (Fig. 22g) using rCDs-

apt1-MoS₂, gCDs-apt2-MoS₂, and bCDs-apt3-MoS₂ as fluorescent monitoring probes for the rapid detection of sulfadimethoxine (SMZ), oxytetracycline (OTC), and chloramphenicol (CAP) in aquaculture. The sensor achieves highly sensitive detection of the three antibiotics within 15 min (Fig. 22h–j), demonstrates high recovery and reproducibility in real shrimp samples, and provides a rapid on-site detection method for food safety. Similarly, Jialu Zhang et al. 211 developed a novel paper-based fluorescent sensor by combining Eu $^{3+}$ -modified carbon dots (CDs) with a sealed paper-based microfluidic chip using a stepwise grafting method (Fig. 22k) for the selective detection of tetracycline (TC) in aquaculture water. The sensor achieves highly sensitive detection of tetracycline (TC) via step-by-step grafting. It exhibits a linear range from 0.1 μ M to 100 μ M and a low detection limit of 0.03 μ M. Additionally, the sensor has demonstrated satisfactory results in real aquaculture water samples, indicating its great potential for on-site detection of environmental pollutants.

Paper-based photodetectors

Paper-based photodetectors utilize the photoconductive effect or photovoltaic effect of semiconductor materials to convert incident light into electrical signals, thereby enabling the detection of light and the transmission of signals. In recent years, paper-based photodetectors have attracted widespread attention and have achieved many research advancements. For example, Shun-Xin Li et al.²¹² used silane modification to form a hydrophobic molecular film on the surface of a paper-based photodetector, creating a highly flexible and deformable paper-based chalcogenide photodetector (PBPPD) (Fig. 23a). The device remained highly stable in humid

conditions, maintaining excellent performance even after 5 days of exposure to 60% relative humidity (Fig. 23b) and under severe bending conditions (Fig. 23c). This work opens up the possibility of producing stabilized paper-based electronic devices. Ting Zhang et al.²¹³ fabricated a paper-based chalcogenide thin film by introducing a seeded layer and using a commercial pen to prepare a chalcogenide photodetector (Fig. 23d). This method solved the difficult problem of preparing paper-based photodetectors and enabled the successful fabrication of large-scale photodetector arrays with high responsivity and image sensors with satisfactory performance (Fig. 23e). The maximum responsivity achieved was 4.2 mA/W, providing an effective strategy for the development of paper-based photodetectors. Cong Zhang et al.214 fabricated a paper-based photodetector by constructing an amorphous indium gallium zinc oxide (a-IGZO)/PbS quantum dots-EDT heterojunction (Fig. 23f). The device based on Senyan paper demonstrated stable detection performance, maintaining its stability even after 1000 bending cycles (Fig. 23g). This approach not only addressed the limitation of quantum dots in paper-based devices due to their low carrier mobility but also achieved significant advancements in the application of wearable electronics. In order to improve the sensitivity and efficiency of the device so that its electrical performance is not affected when bending or stretching, Mehul Dave et al.²¹⁵ fabricated a paper-based photodetector by constructing a heterostructure between WS2 and Ti3C2 Tx. This device exhibits a maximum responsivity of 3.06 mA/W, a specific detectivity of 5.93×10^8 Jones, and a response time on the order of seconds. The responsivity is enhanced by a factor of 10 and demonstrates excellent flexibility and durability, with negligible changes in photoresponse even after extended bending cycles (Fig. 23h). Sunaan Malik et al.²¹⁶ prepared a paper-based photodetector (PD) (Fig. 23i) by depositing and patterning monolayer graphene (SLG), carbon nanotubes (CNTs), and perovskite quantum dots (QDs) to achieve high-performance photodetection within a paper-based system. The fabricated PD achieves an external responsivity of 520 A/W under an operating voltage of <1 V and illumination at 405 nm, exhibiting the highest external responsivity among its peers.

Paper-based supercapacitor

Supercapacitors, as a new type of energy storage device between traditional capacitors and batteries, are considered a promising energy source due to their fast charging and discharging rates, long cycle life, and high power density^{134,217-220}. In recent years, research and applications of supercapacitors as portable energy storage devices have attracted significant attention. Among them, paper-based supercapacitors represent a unique category that integrates supercapacitor technology with paper materials. The working principle of paper-based supercapacitor is similar to that of traditional capacitors, but with higher energy storage capacity. They primarily store energy by forming either double-layer capacitance or Faraday pseudocapacitance on the surface of the electrode material. When a voltage is applied to a paper-based supercapacitor, ions in the electrolyte will form a charge layer on the surface of the electrode material, thereby enabling energy storage. During the discharge process, these ions return to the electrolyte, releasing the stored energy^{135,221}. Paper-based supercapacitors have broad applications across multiple fields due to their unique performance characteristics. For example, in the field of new energy, they can serve as energy storage elements in renewable energy systems such as wind and solar power^{222,223}; in the field of consumer electronics, they can function as backup power sources or fast-charging power sources in devices like smartphones and tablet PCs²²⁴. Additionally, they hold potential applications in the fields of smart packaging and wearable devices²²⁵

In recent years, several studies have been conducted on the development of paper-based supercapacitors. Wanxia Luo et al.²²⁶ employed a filtration method to fabricate a three-dimensional freestanding membrane electrode (BC-E) composed of bacterial cellulose (BC), acetylene black,

and activated carbon (Fig. 24a). This electrode demonstrated enhanced performance, exhibiting excellent mechanical strength, wettability, high electrolyte absorption, and conductivity. The maximum specific capacitance reached 276 F g⁻¹ at a current density of 1 A g⁻¹ (Fig. 24b), with good cycling stability. Under commercial loading conditions, it showed a high specific capacitance of 167 F g⁻¹ and a high energy density of 5.3 Wh kg⁻¹ (Fig. 24c, d). Similarly, Chuanyin Xiong et al.²²⁷ constructed a noncarbonized reduced graphene oxide (RGO)-cellulose nanofiber (CNF) hybrid film (Fig. 24e). This hybrid film exhibited a high specific capacitance of 120 mF cm⁻² (242 F g⁻¹), an energy efficiency of 80%, an energy density of 536 µWh cm⁻² (32 Wh kg⁻¹), and a power density of 193 mW cm⁻² (53 kW kg⁻¹). When directly assembled into a symmetric supercapacitor, it demonstrated excellent mechanical strength and flexibility, outstanding cycling stability (Fig. 24f), and remarkable rate performance (Fig. 24g). Another study by Hao Tang et al.²²⁸ developed a highly conductive composite paper using a simple and rapid vacuum-assisted filtration method (Fig. 24h). The composite paper exhibited superior mechanical properties, with a tensile strength of 34 MPa and a Young's modulus of 6 GPa (Fig. 24i). When the MXene content was only 25 wt%, the composite paper achieved an ultra-high conductivity. The conductivity reached 58,843 S m⁻¹ (Fig. 24j). At a scan rate of 10 mV s⁻¹, a high specific capacitance of 505 F g-1 was achieved in the composite paper electrode (Fig. 24k). Han Q et al.²²⁹ fabricated a foldable, all-paper supercapacitor with excellent self-healing capability by combining screen printing, electropolymerization, and dip-coating methods (Fig. 24l). This supercapacitor exhibited an energy density of 39 $\mu Wh~kg^{-1}$ (69 $kW~kg^{-1}$) and a power density of 692 μ Wh cm⁻² (236 mW cm⁻²), along with excellent selfhealing capability, flexibility, outstanding cycling stability (Fig. 24m), and remarkable rate performance (Fig. 24n). Zhang S et al.²³⁰ developed a supercapacitor using chitosan (CS)/graphene oxide (GO) as the carbon substrate (Fig. 240). This supercapacitor demonstrated an outstanding energy density of 0.585 mWh cm⁻² and a high power density of 3000 mW cm⁻² (Fig. 24p), along with a high specific capacitance of 1050 mF cm⁻² at a current density of 3 mA cm⁻² (Fig. 24q) and excellent cycling stability.

With the advancement of science and technology and the increasing awareness of environmental protection, paper-based supercapacitors, as a green and efficient energy storage solution, hold promising development prospects. In the future, improvements in paper-based supercapacitors will primarily focus on enhancing energy density, reducing costs, and optimizing electrode materials. As related technologies continue to achieve breakthroughs and innovations, paper-based supercapacitors are expected to be widely adopted and popularized in more fields, contributing to the sustainable development of human society.

Paper-based field-effect transistor

Paper-based field-effect transistors (FETs) have demonstrated broad application prospects in the field of flexible electronics due to their unique advantages. With a simple structure and flexible fabrication processes, they can be prepared over large areas through solution processing. These transistors have significant application value in various fields, such as antimicrobial susceptibility testing, flexible displays, and environmental monitoring, making them a research hotspot in recent years. For example, Xin Wang et al.²³¹ fabricated paper-based enhanced-mode complementary metal-oxide-semiconductor (CMOS) carbon nanotube thin-film transistors (Fig. 25a) via a printing process for use in ultra-low-power and radiationresistant flexible electronic devices. These transistors exhibit an extremely low power consumption of as low as $0.0124\,p\text{W}\,\mu\text{m}^{-1}$ and excellent radiation tolerance (Fig. 25b), and are capable of rail-to-rail operation at a supply voltage as low as 0.2 V. Subho Dasgupta et al.²³² prepared paperbased indium oxide nanowire thin-film transistors (TFTs) for wearable and consumer electronics by combining high-throughput solution-processing techniques, including dielectrophoretic alignment and inkjet printing (Fig. 25c). These transistors exhibit excellent performance, including an on/ off ratio exceeding 10⁷, an average linear mobility as high as 42 cm² V⁻¹ s⁻¹,

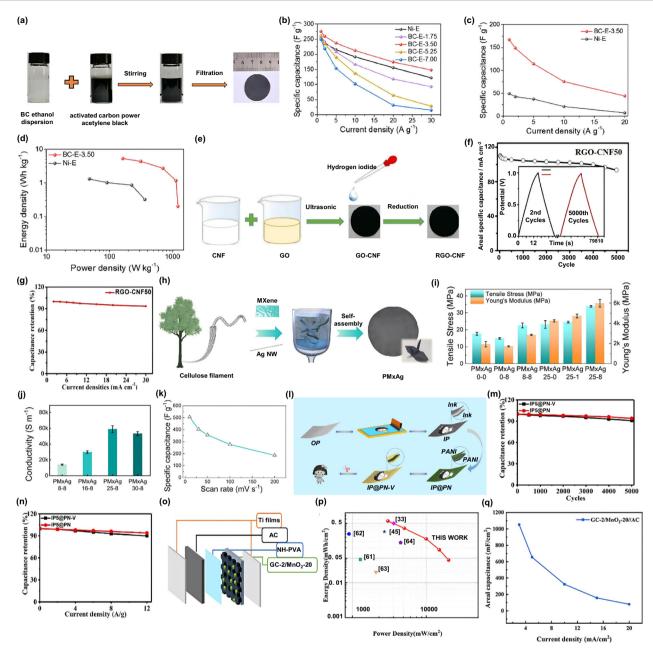


Fig. 24 | **Paper-based supercapacitor and the device performance. a** The preparation procedure of BC-E. **b** Specific capacitance versus current density. **c** Rate performances from 1 to 20 A g $^{-1}$ and (**d**) Ragone plots 226 . **e** The schematic diagram of preparation process of RGO-CNF film. **f** The change of specific capacitance of RGO-CNF hybrid after experiencing 5000 cycles. **g** The capacitance retention of RGO-CNF hybrid under various current densities 227 . **h** Preparation process for composite papers through vacuum filtration and the display of paper bird folded by the produced flexible paper (PM_xAg). **i** Tensile stress and Young's modulus of PM_xAg

composite papers. **j** Electrical conductivities of PM_xAg papers with increased contents of MXene and Ag NWs. **k** The gravimetric specific capacitance calculated at various scan rates of the supercapacitor device 228 . **l** Fabrication procedure of the IP@PN-V hybrid. Comparison of capacitance retention of IP@PN5 and IP@PN5-V hybrid (**m**) at after experiencing 5000 cycles, respectively, and (**n**) various current densities 229 . **o** Schematic illustration of the solid-state asymmetrical supercapacitor device. **p** The Ragone plots. **q** Capacitance retention 230 .

low device-to-device variability, tolerance to extreme stretching strains up to 10%, and outstanding environmental stability (Fig. 25d). Diana Gaspar et al. 233 fabricated dual-gate planar oxide field-effect transistors (DG-FGFETs) on a paper substrate to implement multifunctional logic gate circuits (Fig. 25e). The transistor demonstrates a tunable threshold voltage (VOn) and a high on/off current ratio (IOn/IOff). For a low drain voltage (VDS = 1 V), the IOn/IOff ratio is approximately 8×10^4 , while for VDS = 15 V, the IOn/IOff ratio remains around 4×10^4 (Fig. 25f). Additionally, it can implement universal logic gate functions such as NAND and NOR.

Conclusion and prospects

In this paper, the key elements of paper-based electronic devices are deeply discussed, including detailed analysis of physical and chemical properties of paper substrates, diversified exploration of electrode and active layer preparation methods, and detailed description of the functions and applications of various types of paper-based electronic devices (such as mechanics, electrochemistry, optics, humidity sensors, photodetectors and supercapacitors). We clearly recognize that significant progress has been made in the field of paper-based electronics. The researchers have achieved remarkable results in optimizing material

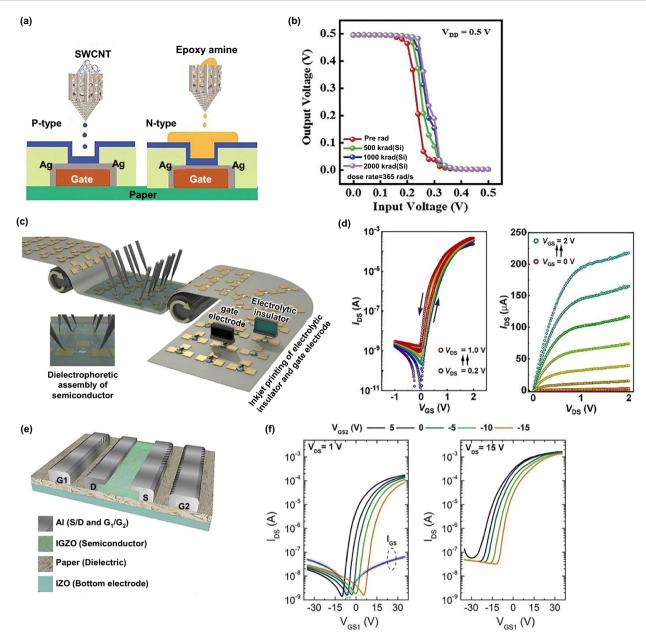


Fig. 25 | **Paper-based field-effect transistor.** a Structure of the P-type and N-type CNT TFTs. **b** VOC curves of a CMOS inverter fabricated with VDD = 0.5 V under different TIDs²³¹. **c** Schematic representation of device fabrication process for large area paper transistors showing each of the fabrication steps. **d** Transfer and output

characteristic curves of paper-based indium oxide nanowire TFTs²³². **e** Schematic illustration of the architecture of IGZO paper DG-FGFETs using the tracing paper as the gate dielectric. f IDS-VGS transfer characteristics of the paper gated IGZO DG-FGFETs at VDS = 1 and 15 V (linear and saturation regime)²³³.

properties and improving device sensitivity and stability, laying the foundation for its initial application in many fields such as wearable devices, environmental monitoring, and biomedical treatment. However, there are still many challenges to overcome. For example, the longterm stability and durability of paper-based materials still need to be greatly improved to meet the actual needs of use in complex environments, the complexity and cost of the preparation process limit largescale industrial production. The integration and multi-function synergies of various devices need to be enhanced to achieve more complex and efficient system applications. Looking forward to the future, with the continuous development of materials science and nanotechnology, it is expected to develop new paper-based materials and functional layer preparation processes with better performance and lower cost. Through deep interdisciplinary integration, further optimize device design and integration, promote paper-based electronic devices to the direction of high sensitivity, high stability, multi-function, miniaturization and intelligence, so as to broaden its wide application in emerging fields such as the Internet of smart home, biosensing and instant diagnosis, open up a new road for the green, flexible and sustainable development of future electronic products, and create more possibilities and opportunities to lead a new round of changes in electronic technology.

Data availability

No datasets were generated or analyzed during the current study.

Received: 2 April 2025; Accepted: 25 June 2025; Published online: 04 August 2025

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Acknowledgements

The authors acknowledge the Natural Science Foundation of China (61935017, 6213000348) for financial support of this work.

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Initial conception: H.S. and W.H. Literature search: W.Y. and C.Z., Writing: W.Y. and C.Z., Guidance: H.S. and W.H. All authors participated in scientific discussion. All the authors read and revised the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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