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Paper: A promising material for human-friendly functional wearableelectronics

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A B S T R A C T

The ever-growing overlap between electronics and wearable technology is driving the demand for utilizing novel materials that are cost-effective, light-weight, eco-friendly, mechanically deformable and can be conformably and comfortably worn on human body as substrate to support the reliable operationof wearable electronic functionalities. Paper materials comprised of bio-origin ingredients (*e.g.,* cellulose and carbon derivatives) have recently attracted remarkably increasing research and commercial interests for prototyping next-generation wearable electronics due to their superiorities including natural abundance, flyweight, mature manufacturing process, specific structural properties, favorable mechanical bendability, biocompatibility and nontoxicity over their counterparts. Feasibility of engaging paper materials has been proved by outstanding performances in body-worn healthcare sensing systems, electro-stimulated artificial muscles, on-site memory storage and wearable power supply on paper substrate. In this review, we present a state-of-the-art introduction of diverse paper substrate options and fabrication techniques employed for realizing paper electronics, and discuss both pros and cons of each manufacturing tactic. Additionally, we summarize developing trends of paper-based electronics in the emerging wearable applications. Based upon these, final conclusions, encountering challenges,accompanied with advancing outlooks are illustrate

Keywords:Wearable electronics Cellulose paper Graphene paper Biomedical monitoring Energy supply

Introduction

Wearable electronics, encompassing a series of smart electronicaccessories and systems that can be conformably and comfortably incorporated on human body, have recently attracted significant attention among labs and markets globally. Examples include flexible displays [1], electronic skins [2,3], healthcare monitoring systems [4–7], energy harvesting devices [8,9], *etc*. The captivating attribute of wearable electronics lies in their capability to fulfill theever-growing demand for instant information interaction [10], real-time physiological function tracking [11,12] and comfortable user experience [13]. The target of enduring less discomfort caused by directly body-worn devices calls for flexible substrate materials.Therefore, the discovery of soft materials is playing a crucial part inthe development of wearable technology. Until now, various soft materials have been used as substrates for wearable electronics. Among these, plastics [14–18] and silicone elastomers [19–22] are the two most commonly employed categories. However, there are several challenges associated with these materials such as the costly production process and adverse impacts on the environment. These factors limit their further applications, particularly those oriented towards large market prospects in underdeveloped regions. For instance, despite the ultra-thin thickness and light weight, plastics, such as PET (polyethylene terephthalate), are hazardous for the environment
[18] because plastic debris is a major source of marine pollution resulting in a rapid decline of global biodiversity [23]. For the silicon-based strategies, such as PDMS (polydimethylsiloxane), apart from the intrinsic non-ecofriendly nature of the material [24], the complex fabrication process that usually requires a time- consuming molding technique [25,26] shows a mismatch with the uprising demand for power-saving and low-cost fabrication schemes.

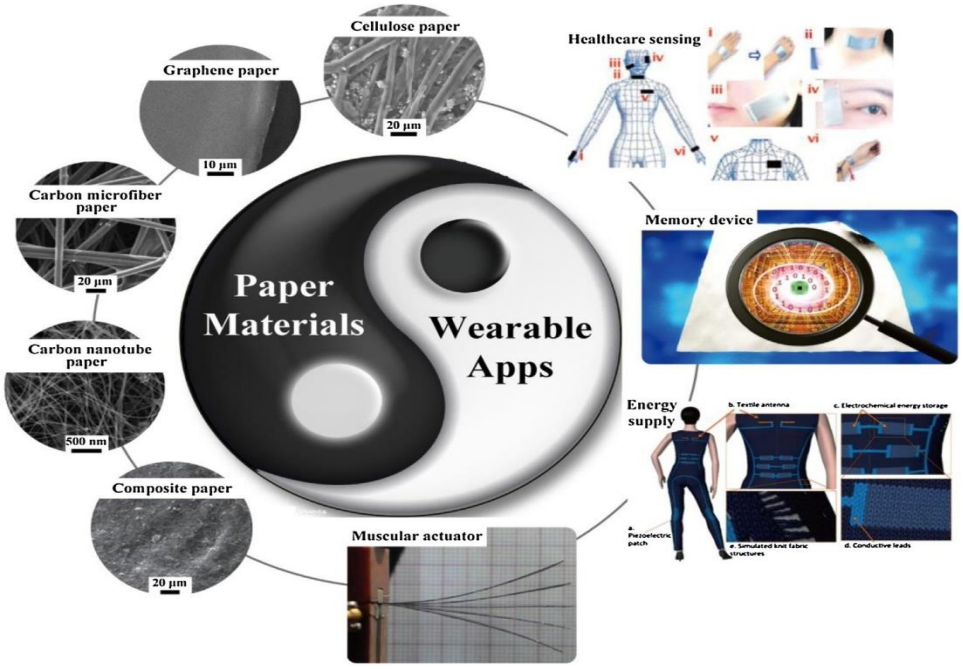

Fig. 1. Paper materials as promising substrates for burgeoning wearable electronics applications. Various paper materials with different compositions (*e.g.,* cellulose, graphene, carbon microfiber, carbon nanotube and composite materials) present a significant potential for applications in the rapidly growing wearable electronics industry for uses as healthcare sensors, memory devices, muscular actuators and energy supply [86,105,130,137,183,187,225,266,267].

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In comparison, paper has been recently unveiled as a promising substrate material for electronics fabrication [27–29] due to its omnipresence, low cost, simple fabrication, green attribute and process-facilitating structure. Paper is ubiquitously available in daily life applications (*e.g.*, writing, packaging, newspapers, currency production, origami art and personal health). Thus, prototyping of electronic components and circuits on paper promises to be meaningful for the potential emergence of multiplexed and multi-functional paper. Besides, the cost-effec- tiveness of paper and the mature papermaking technique pave the way for a rapid, simple and low-cost paper-based electronics fabrication process. In addition, paper is made from raw plant (*e.g.,* wood and cotton) cellulose fibers and thus is recyclable and renewable [30]. Furthermore, paper is capable of serving as a substrate for human-friendly wearable electronics attributing to its decent mechanical deformability and biocompatible property for minimal detrimental effect on human skin. Above all, the porous structure of paper intensifies its superiority over the counterparts in regard to the application in electronic devices. Porous structure is the masterpiece of interwoven cellulose fibers in paper that benefits the coating process, a standard method for enhancing the electrical property of paper by increasing the contact field, which further facilitates the deposition of conductive additives within the intrinsically electrical-insulating cellulose fibers.

Over the past decade, advances in material science have unlocked the research advancement in numerous bio-origin paper materials, all of which are made of naturally abundant ingredients including but not restricted to bacterial cellulose, graphene, carbon microfibers and carbon nanotubes [31–34]. These fresh additions to the paper family show comparable parameters in mechanical flexibility, bio-benignancy and ecological friendliness, as well as some advantages over pulp in making paper electronics. In particular, thin layers of carbon derivatives (*i.e.,* graphene and carbon fibers) are electrically conductive, facilitating or even possibly excluding the conductivity modification procedure, which is crucial and inevitable for prototyping soft electronics on cellulose paper. All these vital properties contribute to arming paper as a series of promising substrate candidates for wearable electronics applications. Wearable electronics have recently witnessed a sharp upward trend of commercialization. Multiple paper materials (*e.g.,* cellu- lose paper, graphene paper, carbon microfiber paper, carbon nanotube paper and composite paper) come to the fore as ideal substrate materials in this realm for using purposes ranging from body-worn healthcare sensing, electronic muscular actuation, memory recording and wearable energy supply (Fig. 1). Although magnificent achievements have been made in paper electronics [35], this novel and rapidly upgrading territory is still confronted with the scarcity of comprehensive reviews reflecting recent advances of electronics that are based on paper substrates and aiming at the cutting-edge wearable applications. In this article, we summarize various substrate selections and methods for prototyping electronics on paper, review recent research progress on application perspectives of paper-based electronics in wearable devices and systems, and finally present several challenges, as well as an outlook on the future development of paper electronics.

1. Substrate selection for paper electronics

Selection of appropriate substrate materials is of crucial strategic importance for accomplishing wearable electronic applications, where soft substrates act as a bridge that connects specific electronic functionalities to devices or garments that can be seamlessly integrated onto human body. In this part, we summarize the production process and discuss physical structure and associated properties of diverse paper materials that are mainly comprised of cellulose paper, graphene paper, carbon fiber paper and composite paper. Additionally, we concisely illustrate potential wearable applications for each material. Based on this information, a material selection diagram focusing on mechanical deformability and electrical conductivity of different paper materials is presented to provide references and guidelines for employing proper materials for paper electronics fabrication (Fig. 2).

Cellulose paper

Cellulose paper has been universally used for a long time with the progress of human civilization mainly for communication and packaging. From the first trials in ancient Egypt dating back to 3000B.C., producing paper with papyrus plants in family workshops, to

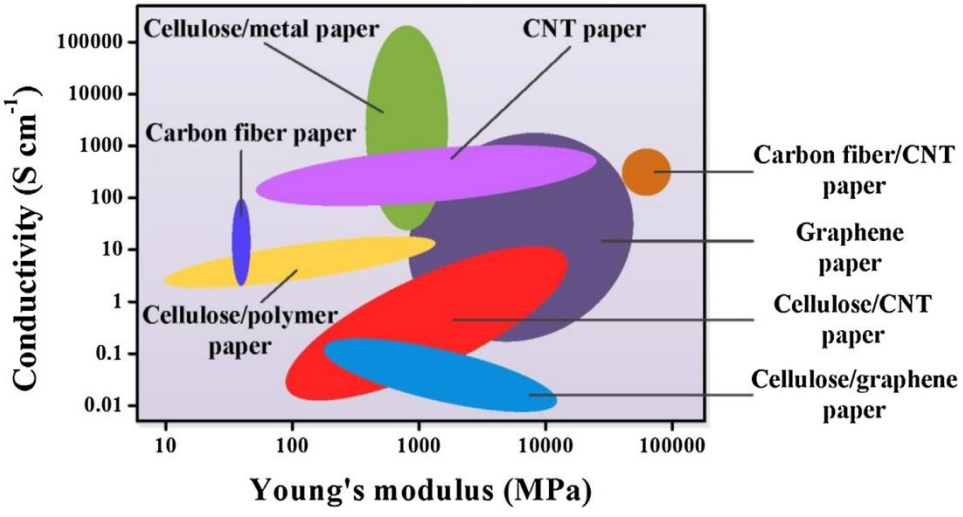

Fig. 2. Material selection diagram: electrical conductivity plotted against Young's modulus. Young's modulus, as a symbol of the mechanical strength, and electrical conductivity, representing the electrically conductive property, are compared among a variety of paper materials. The diagram is helpful to provide design guidelines in materials selection of substrate for fabrication of paper electronics [29,63,75,90,91,102,103,125,160,161,186,267–307].

the intense industrialization of the current paper manufacturing process in modern mills with raw materials named wood pulp, the papermaking industry has

undergone continuously advantageous developments for maturation, mechanization and pipeline opera- tion. The sophisticated papermaking process is initiated with pulping off-cuts such as logs, barks or other cellulose sources with bulk structure to separate cellulose fibers in water suspensions, namely, wood pulps, through mechanical grinding or chemical degrading. After filtration to remove undesired residuals, such as waste from grinding or degrading lignin and hemicellulose, the pulp experiences a beating procedure in which numerous premixes can be added for quality modification depending on the specific purposes of final products. For instance, rosins can improve surface strength, water resistance and thus printability of paper for magazines or gift wrapping, while titanium dioxide benefits opacity and brightness enhancement of paper. In the subsequent process, dilute pulp solution is subjected to repeated mesh screening, roller pressing and vacuum drying to drain off water and finally transforms into the actual sheets of paper. Further, considering various ultimate applications of paper, extra operations including smoothing, compacting and coating may occur before the product is available for family, industry or research uses.

Cellulose paper is generally known to possess a porous structure with cellulose fibers connected via hydrogen bonds. This porous internal architecture can bring about dramatic convenience for paper modification because the interconnected cellulose network increases the contact area and provides sufficient binding sites for depositing additives through which paper can be tuned with specific attributes and functionalities. Regarding the paper electronics application, such porous construct boosts the deposition of conductive fillers and accordingly alters the inherently insulating paper to be electrically conductive. Additionally, natural fibrous cellulose can be further divided into thousands of repeating constituent units named cellulose nano- fibers [36–38]. These cellulose fibers have a nanometer diameter, which is much lower than the wavelength of visible light. Thus, paper that is directly composed of cellulose nanofibers is optically transparent. Moreover, this nanopaper can be tailored from clear to opaque by changing fiber size and porosity. These endeavors are extraordinarily meaningful for wearable applications especially where optical transparency is significantly favored such as in electronic paper displays and solar cells for wearable energy conversion/supply.

Recently, bacterial cellulose (BC), a unique type of natural cellulose produced by certain species of bacteria, has emerged as an interesting material due to its better biocompatibility, higher tensile strength, and increased water retention ability compared with plant cellulose [39]. BCs usually form pellicles consisting of interconnected ultrafine ribbon-like nanofiber networks, and the BC fibers are of premier chemical purity and do not contain lignin or hemicellulose [39]. Besides, one hallmark of BC is that the BC fibers are significantly smaller than those of wood cellulose, and the porosity of the BC pellicle (or BC paper) can be tuned with different fiber densities [40], easing the process of electrical conductivity enhancement where suitable porosity is appreciated. Cellulose paper, as the main representative of the paper family, is highly anticipated in wearable electronics areas to serve as a versatile substrate due to its light weight, bio-friendliness, mechanical bendability and unique material structure that facilitates the electrical quality improvement process. Meanwhile, the low cost and simple process of fabricating paper-based electronics, such as pen writing techniques [41], further suggest that cellulose paper is practically feasible for this application. From an additional perspective of environmental protection and sustainable development, cellulose paper is also an optimal substrate choice for wearable electronics. More specifically, it is clearly witnessed that swift progress in science and technology is currently propelling the upgrade and replacement of electronic products in consumer market, which in other words implies the decreased lifecycle of consumer electronics and correspondingly increased consumption of natural materials. Paper comprised of cellulose fibers can be degraded by fungi in soil, and it is a naturally renewable material, providing a potentially viable solution for the environmental concerns. Until now, cellulose paper has surpris- ingly created a significant interest that embraces most electronics fields ranging from basic electronic components (e.g., conductors, diodes and transistors) [42–47] to functional devices and systems [47–52], and this interest is still prominent.

Graphene paper

Graphene paper broadly refers to graphene-based paper, which includes graphene oxide (GO) paper [53], reduced graphene oxide (rGO) paper [54] and functional graphene paper [55]. GO paper is intrinsically a type of micrometer thick membrane material fabricated from GO [53]. Normally, GO is electrically insulating due to the scattering effects of oxygen functional groups distributed on its basal planes and edges. However, its electrical property can be improved by chemical reduction or heating to remove the oxygen functional groups, in other words, to transform it into an rGO paper [54,55]. This type of rGO paper has many technological applications in body-worn electro-active artificial muscles [56,57] and wearable energy supply [58–60] due to its peculiar electrical transport property, high thermal stability and exceptional mechanical strength.

Graphite is a raw material for graphene paper production. First, it undergoes a chemical exfoliation process to be synthesized into GO nanosheets. This chemical exfoliation is a liquid-phase approach named the improved Hummers method [61]. The method generally includes three procedures: 1) oxidation of graphite by introducing strong acids and oxidants, 2) exfoliation of graphite layers via sonication, and 3) purification of exfoliated flakes from un-exfoliated forms using an ultracentrifugation method. Subsequently, the semi-finished nanosheets can be prepared into graphene paper via a variety of ways, mainly including a vacuum filtration method [53,54,62] and a freezing- drying and pressing method [63]. The vacuum filtration method is the most popular route among them. In the operation, GO nanosheets are dispersed in water or other solutions with the aid of an ultrasound cleaning bath to form a homogeneous colloidal dispersion of a certain concentration. Then, the colloidal dispersions are vacuum filtered through an Anodisc membrane filter, followed by successive steps involving washing, air drying and peeling from the membrane filter to yield GO paper. This fabrication approach is simple, inexpensive and therefore exten- sively used in the preparation of graphene and its derivative paper. The freezing-drying and pressing method [63] resembles the vacuum filtration strategy in the first step. The difference is in the subsequent procedure of freezing-drying to fabricate the GO aerogel followed by thermal reduction and pressing of the intermediate product to eventually obtain graphene paper. This methodology can produce graphene paper with a folded structure that may be suitable for expanding the space between adjacent graphene sheets and then used for enhancing reversible capacity of battery anodes. However, the abovementioned approaches are either a simple lamination of disordered sheets [63] or a semi- ordered accumulation [53,54,62], which hinders the accurate control of the process to ensure the highly ordered structures that are of critical importance to mechanical strength improvement and interfacial conductivity enhancement.

Thanks to the excellent performance in electron transport and mechanical strength, graphene paper, mainly rGO paper, has found diverse wearable applications. The latest attentions have been paid to body-worn power supply, explicitly as anodes or cathodes for energy storage devices [64,65]. Increasing efforts have been made to further enhance its performance by introducing functionalized additives to graphene paper [66–69]. Low electrical conductivity of pristine graphene paper (200 S Cm—1) has set a barrier for its applications as electrodes for a long time. To settle this, highly conductive silver nanowires were doped onto graphene paper to dramatically improve its electrical conductivity (3189 S Cm—1) [68]. Apart from providing energy solutions, graphene paper has also been exploited in actuation studies, presenting spontaneous mechanical response to external electric stimuli, which is highly suitable for wearable artificial muscles [70]. In addition, unreduced GO paper can be functionalized to be used in biomedical applications because of the abundant existence of functional groups such as carbonyl, hydroxyl and epoxide groups in GO [71– 74], which is of extraordinary value for on-site health monitoring through electrochemical detection of physiological signs.

Carbon fiber paper

Carbon fiber paper (CFP) is a flat sheet of microscale carbon fibers with porous structure and attractive properties including large specific surface area, medium electrical conductivity (100 S Cm—1 [75]) and the potential for cost-effective mass manufactur- ing. During the past decade, CFPs have experienced extensive studies for use as conductive electrodes and substrate matrices of lithium ion batteries [76–78], fuel cells [79] and supercapacitors [80–84], and were determined to be practical for energy solutions for wearable electronics. These remarkable applications in various energy storage devices rely on the fundamental principle of the pore structure of CFP. In concrete terms, pore size and distribution can significantly impact the operating performance of electro- chemical capacitors [85]. Additionally, the pristine porous network capacitates CFP to serve as

the electrode/electrolyte interlayer substrate for lithium ion batteries and fuel cells, mainly by providing pathways for ions and electrons and by trapping dissolved electrolyte materials without the charge transport limitation [78].

To further reinforce the electrochemical performance of energy storage devices made of CFPs, multiple metallic coatings on CFPs have been carried out. For example, gold-modified CFPs with enhanced electrical conductivity in comparison with raw CFPs have been used as anode materials of fuel cells [79]. Nickel/cobalt- based nanomaterials with different compositions and morpholo- gies (e.g., nanosheets [81,83], nanotubes [82], nanowires [83] and nanonets [84]) have been grown on CFP's surface, and the obtained Ni/Co/CFP composites exhibit improved redox kinetics for pseudocapacitors as induced by the three-dimensional (3D) hierarchical architectures and the short diffusion path for swift ion/electron percolation after adding the Ni/Co nanocomposites. As a result, such modifications resulted in corresponding improve- ments in rate capability (scan rate as high as 100 mV s—1 [84]) and cycling stability (excellent performance even after 5000 cycles [84]), which bring CFPs a step closer to practical energy storage uses.

Recently, a thin film of hollow nanosized carbon fiber aggregate,
namely, carbon nanotube (CNT) paper, has been used to supply energy to wearable electronics [86–89] due to its light weight (0.04 mg Cm—2 [89]), superb electrical conductivity (2000 S Cm—1 [90]), strong mechanical robustness (Young's modulus of 25 GPa [91]) and porous construct with high aspect ratio which can benefit ready charge transport. However, common CNTs with a one- dimensional structure have limited capacities (maximum value
less than 700 mA H g—1 [92–94]). To resolve this, research was conducted to combine CNT paper with highly capacitive materials [95,96], but the enhancement effect is unfortunately not pro- nounced. Another tactic is to alter the primitive geometrical construct to 3D interpenetrated CNT forests via the floating catalyst approach [97] or simply by stacking CNT sheets together [98]. Using these approaches, a surprisingly high specific capacity of 34,600 mA H g—1 under the current density of 500 mA g—1 was obtained [97]. Apart from the aforementioned features, CNT paper is also applicable for efficient heat dissipation in electronic devices because CNTs are well known for their thermal conductivity [99– 101]. Specifically, CNT paper may protect electronic circuits from overheating under high power and long-term operation, and thus may lead to more advances in miniaturized wearable electronics.

Composite paper

Composite paper refers to a series of paper-like hybrids with independent pairs of cellulose, graphene, carbon microfibers or CNTs. Cellulose-based composite papers have attracted significant attention as substrates for wearable electronics because of the abundance of cellulose in nature, straightforward manufacturing process of these composites and, consequently, a relatively low expense for production and applications. However, there is a theoretical hurdle for blending cellulose with some carbon derivatives, especially CNTs, because they are hydrophobic while fibrous cellulose is hydrophilic, and CNTs tend to aggregate, which impedes the uniform dispersion of CNTs in the cellulose construct. To circumvent this, anionic surfactant was employed to modify the CNT surface with negative charges [102,103]. Because cellulose fibers are also negatively charged, CNTs can then bind to cellulose via ion bridging of a cationic fixer, and this fixer blocks the dispersed CNTs from aggregating. Thus, the electrically conductive cellulose/CNTs composite paper with a specific conductance of
671 S m—1 can be produced via the conventional papermaking
method [102], and it shows prospect to be used as fundamental unit in various wearable electronic systems.

In addition to CNTs, cellulose is also used with another conductive carbon allotrope (i.e., graphene) to form a composite paper that shows potential for uses in electronics [104–107]. Graphene has long been known as a dominant electrode material for energy storage devices ever since its discovery [108–110]. The interaction with cellulose is theoretically possible to further optimize the performance of graphene. The addition of cellulose can provide a porous network architecture for the growth of graphene, where graphene can subsequently be fixed to the cellulose fiber via strong interactive bonds and fill the pore and further cover the entire pore texture of the cellulose net-structure [105,106]. Thus, the surface area of graphene can be increased, which leads to improved specific capacity and reversible capacity retention [105,106]. In addition, due to the blend with a mechanically robust fibrous cellulose, the cellulose/graphene composite paper presents enhancement in mechanical strength, and therefore outperforms the graphene-only film in long-term cycling stability when used in a power supply for wearable electronics [105,106].

It is worth noting that composite papers of pure carbon
materials, such as the carbon microfiber/CNTs composite [111–113] and graphene/CNTs composite [114,115], are also being tested for applications in wearable energy supply. In one case, freestanding graphene/CNTs hybrid paper was produced via vacuum-filtration of water suspensions of graphene nanosheets and CNTs, aiming to increase the cross-sectional conductivity and specific capacity of the composite paper through the adoption of CNTs, and further prompt its potential for battery uses [115].

2. Various techniques for fabricating paper electronics

Coating of paper electronics

Coating has long been a classical and industrially mature tactic for material surface modification via layer-by-layer deposition, through which the modified materials are made to satisfy a variety of appealing applications, including retrofitting in electronics [116–118]. Ever since the proposal of the next-generation paper electronics concept, the sophisticated coating technique has emerged to realize the electrical conductivity refinement of paper by adding conductive fillers onto the surface or into the inner fibrous structure of such materials [104,119,120].

A large number of coating techniques, *i.e.*, Meyer rod coating [119], dip-coating [121], spin-coating [122,123], sputtering
[122,124], soaking [125,126] and evaporation [127–129], have revealed the compatibility with functionalized electronics pro- duction on a paper base. In one case, an old but stable Meyer rod coating method was used on Xerox paper to convert it into a super conductive material (sheet electric resistance of approximately
1 Ω sq^{-1}). The obtained highly conductive paper was further
adopted to prototype energy storage devices with an excellent cycling stability (less than 3% of capacitance reduction after 40,000 working cycles [119]). By focusing on energy solutions, the collaboration of spin-coating and evaporating techniques resulted in the fabrication of a paper-based nanogenerator (Fig. 3a) [123], in which the evaporated silver (Fig. 3b) and spin-coated poly-tetrafluoroethylene (PTFE) (Fig. 3c,d) layers on paper operated under the electrostatic effect to convert mechanical deformation to electricity [123]. Another instance suggests that simple deposition of silicon nanoparticles onto microfibers within CFPs can efficiently improve the overall capacity, hence making CFP a more reliable anode material option for wearable energy supply [130]. This series of layer-by-layer coating techniques is widely favored because these techniques usually provide a desirable smooth and homogenous surface structure after coating and a well-tailored thickness of the coated additives (nano-size to micro-scale [104,121,123]) by adjusting the coating period and cycles, both of which are advantageous for obtaining paper electronics with tunable performance. Another positive feature of coating strategies is the abundant selection of coating additives. So far, as we investigate, conductive additive materials with distinct physical and chemical properties, involving metals [119,121,122,124,126,128,129,131], carbon-derived nanomaterials [104,105,119] and conductive polymers [125,127,132], have been used for coating paper electronics with specific functionalities. For example, indium-zinc-oxide and gallium-indium-zinc-oxide films were, respectively, coated on either side of the cellulose nanopaper via the sputtering process to establish an N-channel of a paper- based transistor that can be used in power-saving circuits [124]. Combined with newly synthesized magnetic electrophoretic particles, TiO_2 particles were evaporated onto the paper to obtain a bimodally driven electronic display [128]. This electronic paper display exhibited a satisfactory response to both electric and magnetic fields [128], which makes it potentially beneficial for developing identification tags and security labels.

However, coating of electronics on paper is also undeniably
facing some limitations. Commonly, the coating techniques do well in depositing layers of materials onto substrates with tunable vertical

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resolution but struggle in patterning specific geometries, which severely prohibits their further prospect in electronic applications where products with precise geometrical constructs and accurate spatial resolution are generally needed. Although the attempts of integrating a shadow mask [124] present a possible solution, the extra-introduced cost and procedure may tarnish the reputation of being easily accessible. Additionally, the use of vacuum conditions [104,131] for some coating methods presents the same challenge in ramping up manufacturing expenditure and complexity.

Printing of paper electronics

Printing of electronics has become an increasingly popular technique for electronics industry due to its superior properties such as low cost, rapid prototyping and potential for widespread applications compared with conventional coating strategies [133– 135]. It is well known that paper is a ubiquitous printed media in industry, education and daily life. Recently, thanks to the rapid development of printing technologies and significant advance- ment in paper making industry, the integration of printing method and paper substrate has drawn tremendous research interests in fabricating electronics with reduced cost, enhanced adaptability and enriched functionality [44,136,137]. The underlying mechanism of such paradigm is simply patterning conductive traces on

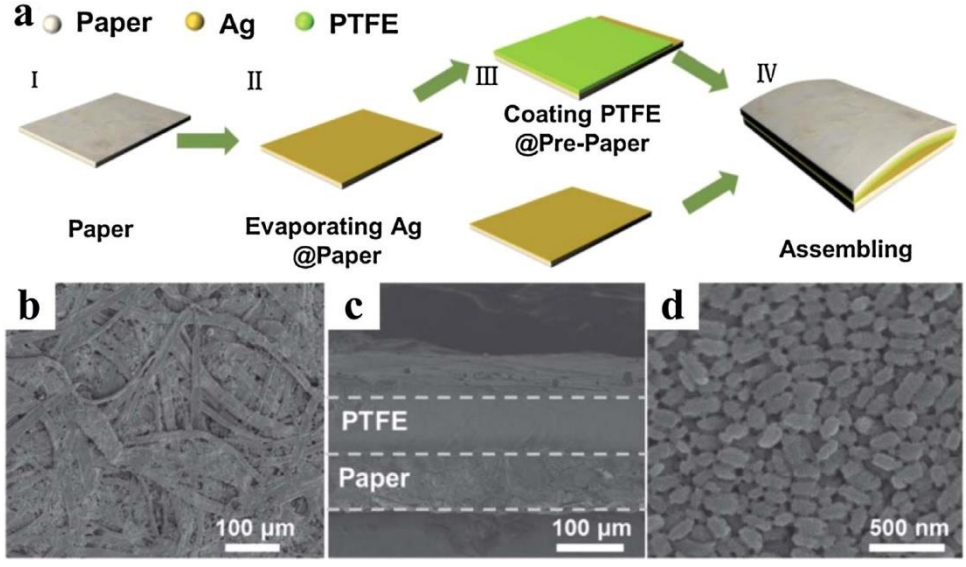

Fig. 3. Paper electronics fabricated by coating techniques. (a) Schematic illustration of the coating process. (b–d) Scanning electron microscopy (SEM) images of paper evaporated with silver (b), cross-sectional (c) and top (d) view of paper with successive evaporation of silver and spin-coating of PTFE layers [123].

paper surface or dispersing electronic inks into its porous structure for electrical conductivity enhancement via printing procedures. Therefore, the selection of a proper combination of printing approaches and conductive inks becomes essentially crucial for practicably printing electronics on a paper sheet.

From the past to the present, from lab to industry, many ingenious printing strategies have been brought to the scientific and technological frontline of printing electronics. Inkjet printing, as a commercially available technique, has achieved the most extensive industrial and daily applications due to its micro-scale printing resolution [29,137]. Originating from ancient China more than 2000 years ago, screen printing has found its applicable prospect in wireless communication, and it is employed for creating antennas on a nanopaper composite [44]. These sheet-processing techniques reveal a satisfactory suitability for lab-scale applications. However, these techniques have a low throughput and a limited industrialization potential. Gravure printing, a typical Roll to Roll (R2R)-based tactic, is suggested as a large-volume manufacturing approach for its high printing speed (20–1000 m min^{-1} [35]) versus other printing techniques. In addition, the R2R printing technique has emerged as a workable approach for producing photovoltaic cells on paper (Fig. 4a,b) with homogenous surface morphology (Fig. 4c,d) and distinguishable layer structure, which is mainly comprised of paper substrate (Fig. 4e), zinc cathode, photoactive layer and conductive anode (Fig. 4f) [138].

Ink selection for such printing scenarios significantly varies. The inks can be based on conventional metallic materials (*e.g.*, silver [29,30,44,136,137] and zinc [138]), emerging carbon materials (*e.g.*, carbon paste [137] and graphene [139]) and organic conductors (*e.g.*, conductive polymers [138]) to meet diverse application purposes. For instance, micro silver powders with excellent electrical conductivity and mechanical robustness were screen- printed onto commercial printing papers to form conductive patterns for paper-based printing circuit boards (P-PCBs) [30]. With surface-mountable functional components directly integrat- ed along the patterned conductive traces, the multi-layered P-PCB was suitable for accomplishing some complicated electronic processing tasks. In addition, an ISO-guided life cycle impact assessment implied that compared with popular organic silicon- based PCBs, P-PCBs had a far lower impact on our ambient environment as indicated by a significantly lower environmental impact index [30], which facilitates the upcoming green electron- ics, including those for wearable applications. Apart from common conductive inks, photonic ink [140–142] has also unveiled its

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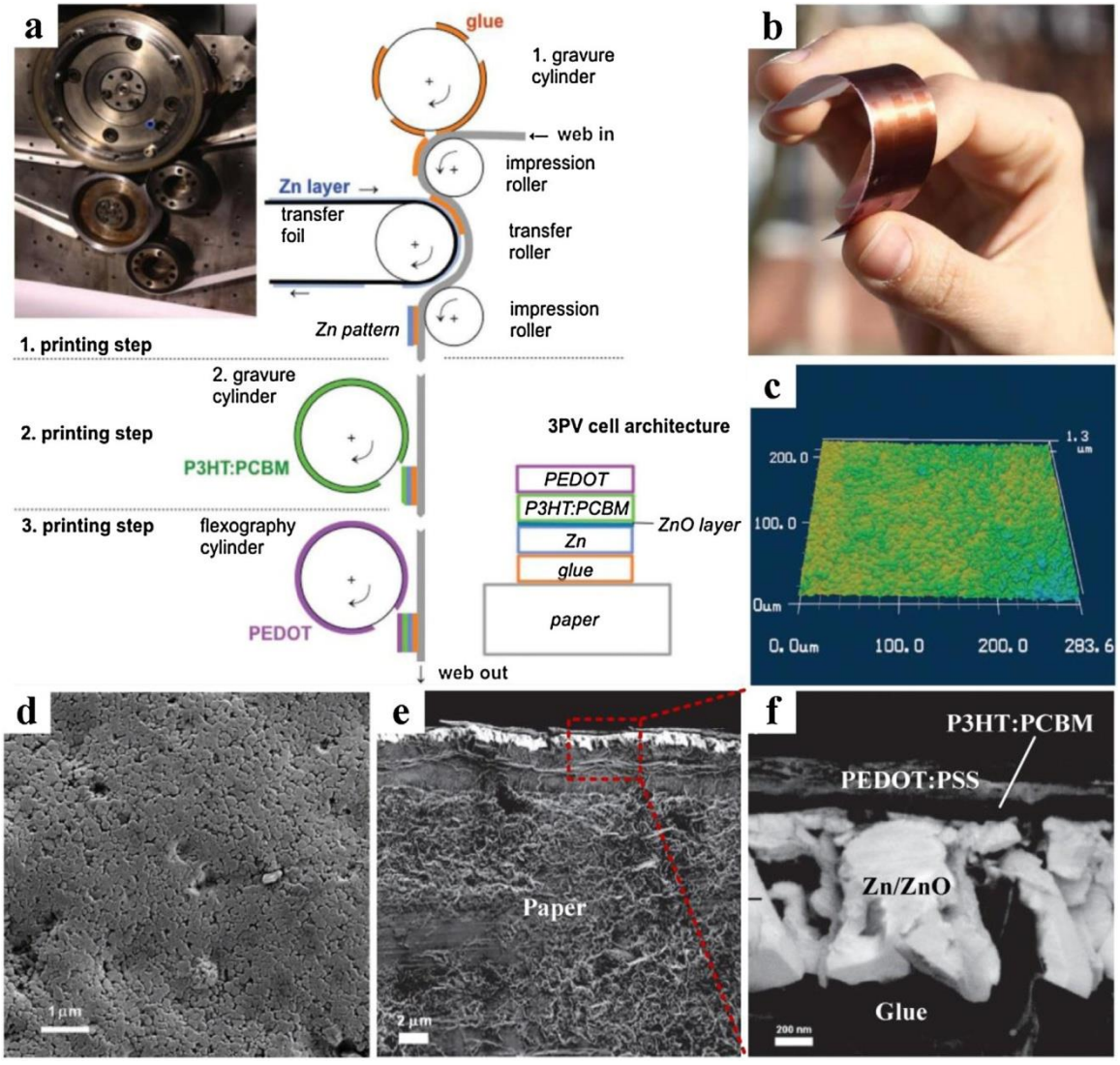

Fig. 4. Paper electronics fabricated by printing techniques. (a) Optical image and schematic diagram of the printing process. (b) Digital photograph of the printed photovoltaic cell. (c, d) Laser scanning image (c) and SEM image (d) of the surface morphology of the fabricated cell. (e, f) Cross-sectional SEM images of paper-based cell at lower (e) and higher (f) magnifications [138].

promising application in printing paper electronics, such as paper- based photoelectric display. The photonic ink is based on photonic crystals, and via engineering the photonic band gap of such materials, the wavelength of reflected light can be controlled to realize unique photonic phenomena. Under this mechanism, photonic ink can display any color in the optical spectrum, prompting the development of full-color photoelectric displays [143–145].

The mature development of printing industry promises its technical advantage in rapid prototyping and ease of scaling up for producing paper electronics. Nevertheless, some challenging issues are demanding effective solutions before the P-PCBs scenario can move forward as a low-cost, user-friendly approach. One problem results from the necessary engagement of bulky, professional instruments, which increases the manufacturing cost through purchasing a printer from the market or establishing a printing system in lab. Even though inkjet printing dominates the largest share in printing industry, the accurate selection of ink materials is still threatening its widespread applications in fabricating paper electronics. Furthermore, because small nozzles are often employed for high-resolution printing, the conductive inks for inkjet printing are required to be of properly low viscosity and low evaporation rate to avoid clogging. However, the printing speed that these techniques have always been known for is unexpectedly restricted by the long drying period of the slowly evaporating inks.

Writing of paper electronics

Recently, inspired by day-to-day hand writing, scientists and researchers have developed writing techniques for fabricating paper electronics using commercially available pens [41]. Com- pared with the abovementioned techniques, writing techniques show specific advantages including easy accessibility, rapid prototyping, low cost, as well as user friendliness. Particularly,"on-demand" wearable sensors catering for "on-site" purposes can be easily fabricated by the end-users using pen-based writing techniques [146].

Various types of pens have been developed to fabricate paper electronics such as a brush pen [147–149], pencil [150–153], fountain pen

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[154,155] and ball pen [156–159]. Russo et al. first developed a silver ink filled ball pen to directly write conductive silver interconnects on paper (Fig. 5a,b) [156]. Silver ink is specially designed with viscosities of 1–10 Pa s to make the ink smoothly flow through the ball pen tip during writing. In addition, the copper nanoparticle ink was synthesized for directly writing flexible electronics on paper using a ball pen [160,161]. Further- more, the ubiquitous ball pen has also found applications in fabricating plasmonic papers [162,163]. In this bioplasmonic calligraphy strategy, bio-functionalized gold nanorods were patterned onto a piece of filter paper to establish an inexpensive, highly sensitive and facile platform for multiplexed bioassays and selective chemical detections, facilitating its application for diagnostic purposes in some resource-limited areas. More recently, an enzymatic ink-based ball pen was proposed to directly write electrochemical biosensors with good bio-catalytic activity and electrical conductivity, eliminating the enzyme modification process on the electrodes [146]. Particularly, the ball pen filled with glucose oxidase-based ink was successfully employed to directly write a glucose sensor on human skin for self-testing of glucose, revealing a promising prospect for household personal- ized healthcare. Notably, the ink preparation process is of great significance for direct writing electronics using ball pen to prevent leaking and clogging, which, however, is time-consuming. To address this difficulty, our group have created a pressure-assisted ball pen methodology to write electrically conductive carbon and silver electrodes on paper with no requirement for excessive ink preparation, showing universal compatibility with numerous working conditions [164].

Different from the metal particle-based inks, the room temperature liquid metal ink shows attractive features, involving low-melting points, high electrical conductivity, well-defined biocompatibility and well-controlled wettability. Liu's group for the first time directly wrote gallium-based liquid metal ink on paper under room temperature using a brush pen for flexible paper electronics fabrication (Fig. 5c,d) [148]. In addition, several functional electronic components, such as inductors, capacitors and resistors with satisfactory performance, have been demon- strated by directly writing liquid metal on paper [149]. Because the liquid metal ink remains in liquid phase at room temperature, the patterned electronics using this material has a self-healing property, which ensures a swift recovery of conductivity even

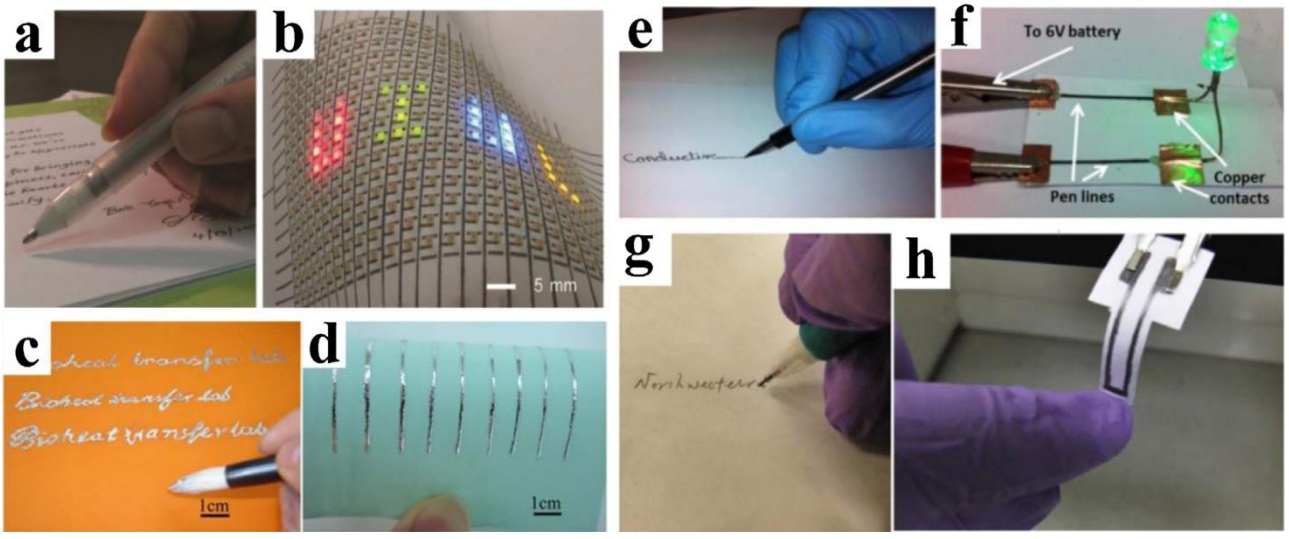

Fig. 5. Paper electronics fabricated by writing techniques. (a, b) Digital photographs of electronics fabrication via ball pen writing (a) and a paper-based LED display (b) [156]. (c, d) Photographs of writing conductive text (c) and lines (d) on paper by a brush pen [148]. (e, f) Images of the fountain pen writing process on paper (e) and function demonstration of an LED powered by a power source using the written electrodes on paper (f) [155]. (g, h) Images of the pencil writing process on paper (g) and a U-shaped pencil trace written on a paper acting as a strain gauge (h) [28].

after the conductive patterns are broken. Hence, this liquid metal ink accompanied with brush pen is promising for writing functional wearable electronics.

In addition, fountain pens loaded with carbon-based inks have also been developed for direct writing of electronics. For instance, carbon nanotube ink [154] and carbon nanofiber ink [155] were successfully synthesized and applied for writing conductive paper electrodes using a fountain pen, provoking the development of paper electronics (Fig. 5e,f).

As stated above, the writing strategies that use a brush pen, fountain pen and ball pen are the typical solution processing techniques and thus need to be modified with a suitable conductive ink prior to writing. In comparison, writing with pencil is a solvent-free technique and thus can be used to directly write carbon-based electronics due to the good electrical conductivity of graphite. Pencils with diverse degrees of hardness (H) and blackness (B) have revealed feasibility for writing electronics on paper. For instance, 4 B pencils have been used to produce a photodetector [165] and UV sensors on paper [166,167], while 3 B pencils have been used to prepare carbon electrodes on paper for configuring electrochemical sensors [168,169]. In particular, because the pencil trace written on paper is made of graphite particles in close contact, the resistance of the pencil trace will change subjected to the substrate deformation (*e.g.,* tension or compression) depending on the degree of contact between the adjacent particles [150]. Based on this principle, the pencil-written trace on paper has been utilized to act as a sensing element for graphite-based strain sensors (Fig. 5g,h) [150,170,171]. More recently, bio-inspired electronic whiskers based on writing conductive graphite traces on paper have been explored for tactile sensing application, indicating potential applications in artificial intelligence and human health monitoring [172].

Writing electronics on paper using ubiquitous pens has been

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proven to be a promising technique for fabricating paper electronics due to its advantages of being simple, inexpensive, rapid and portable. Brush pen, fountain pen and ball pen are compatible with multiple conductive inks for making both carbon- based electronics and metal-based electronics, including the highly conductive gallium-based liquid metal [148,157], copper ink [160,161,173], silver ink [174–176], carbon nanotube ink [177]and enzymatic ink [146]. Whereas the pencil writing is well adaptable for making carbon-based electronics on paper in a solvent-free manner [151]. However, for this technique, the writingspeed and the repeated writing cycle need to be improved during the writing process. Additionally, mass production of paper electronics using this writing technique is still a challenge.

Microfluidics fabrication of paper electronics

The previously introduced approaches are primarily used for patterning conductive circuits on the paper substrate surface. Although these strategies provide high spatial resolution, they have several limitations. Specifically, 1) poor electro-mechanical stability because the coating layer does not robustly adhere topaper, and detachment and cracks may occur on the conductive layer after folding and bending, which poses a limitation for the industrialization of paper electronics. 2) Difficulty of fabricating multilayer inter-connected electronics without cutting holes and complicated alignment. Microfluidic devices have been widely investigated for wearable and flexible electronics [178–180], where microfluidic channels provide a strong support for conductive materials and better electro-mechanical stability compared with directly patterning circuits on surfaces. Incorporation of micro- fluidic and electronic elements into one system, namely, micro- fluidic electronics, opens possibilities for new functions and devices for wearable technologies, such as micrototal analysis systems. Today, most devices employ PDMS as a substrate due to itsrelatively mature fabrication technology, stable and elastic

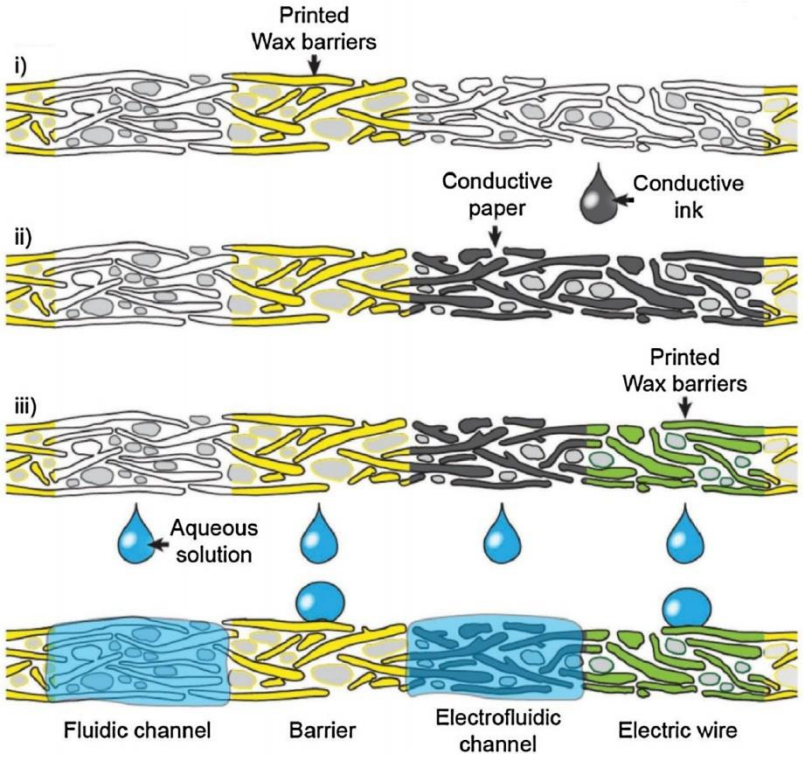

Fig. 6. Paper electronics fabricated by microfluidics techniques: with the help of wax barriers. 1) Printing wax on paper to form a channel for conductive materials inside the paper. 2) Adding inks to the wax-defined channels. 3) Another round of wax printing to further confine the liquid microfluidic channel and fix the conductive network.

mechanical properties, as well as repellence of water solution. Because the paper substrate is naturally porous and permeable to most liquids, it is challenging to use it for microfluidic-based electronics.

To overcome the challenges, one solution is to expand the third dimension of paper electronics from the surface to inside the paperusing water-dispersed conductive inks [181]. The device fabrica- tion process includes three main steps (Fig. 6). Specifically, 1) printing of wax on paper and melting it to form a channel that defines the structures of electronic materials inside the paper. 2) Addition of inks to the wax-defined channels. Driven by capillary wicking, the inks spread along the channel. After drying the ink, the paper fibers are coated with inks and form conducting networks inside the paper that are defined by the channel. 3) Another round of wax printing is used to further confine the liquid microfluidic channel and fix the conductive network. In contrast with the abovementioned coating methods, the inks used here are supposed to be: 1) less viscous and able to wick in the paper substrate and easily dried and 2) of the particle size (if the ink contains particles) smaller than the paper pore size but large enough to form a network after drying.

Another promising approach for solving the problems associ- ated with the surface coating method is to develop paper/polymer composites

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[182] that render paper hydrophobic and lay the foundation for paper-based microfluidic electronics. Paper sub- strate was etched using a user-friendly laser cutting system, and then stacked to form a 3D physical microchannel. Finally, the entire paper-based device was immersed into a polymer liquid to block the porous structure of paper. Based on this method, a 3D flexible paper conductive wire was designed and realized as a light switch. More recently, Liu et al. systematically investigated the interaction of paper and liquid metal [183], and surprisingly found that the porous cellulose paper substrate (Fig. 7a) is metallophobic (Fig. 7b) and impermeable to liquid metal (Fig. 7c). This significantly simplifies the fabrication technology (Fig. 7d) because paper does not need any extra modification, and the corresponding results showed a favorable patterning resolution (Fig. 7e), feasibility of 3D construction (Fig. 7f) and appealing electrical stability of paper conductors even after 1000 cycles of bending. Additionally, the authors are the first to show the concept of a paper-based pressure sensor that moves paper electronics forward to practical applica- tions.

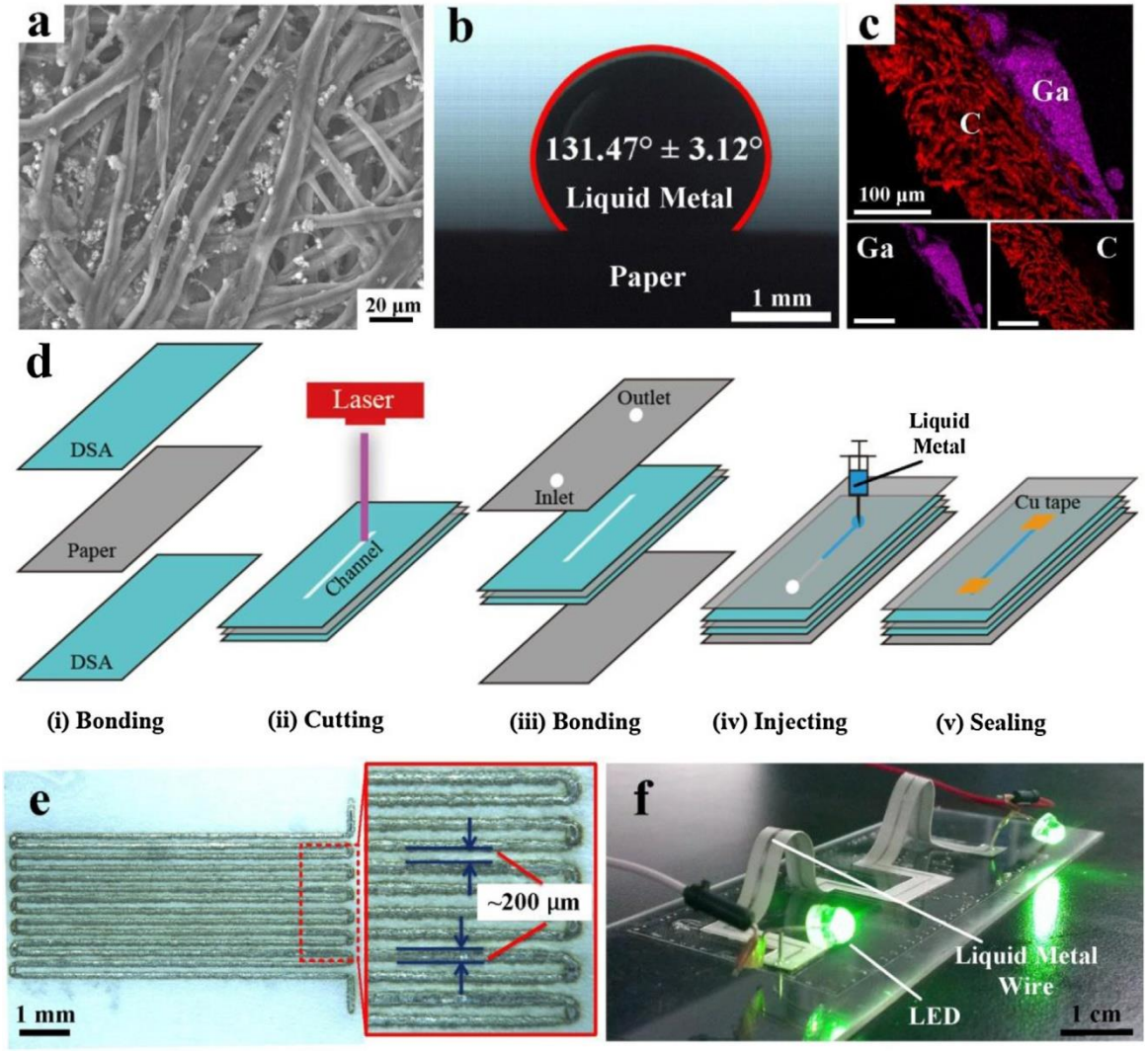

Fig. 7. Paper electronics fabricated via microfluidics techniques: direct etching of channels on paper. (a) SEM image of the surface structure of paper. (b) Contact angle of liquid metal on the paper substrate. (c) Energy dispersive spectrometry (EDS) elemental analysis of the paper in cross-sectional view to observe liquid metal penetration into porous paper substrate. (d) Schematic of the microfluidics fabrication of paper electronics. (e) Optical image of liquid metal conductive lines with a spatial resolution of approximately 200 μm (inset). (f) Digital photograph of a 3D bridge-shaped conductor [183].

3. Emerging wearable applications of paper electronics

Healthcare sensors

Provoked by the emergence of a novel personalized medicine trend that demands showcasing individuals themselves with insightful information about their health status, wearable elec- tronic products are timely engineered to promise optimal operation of this healthcare industry. In this section, we present state-of-the-art research advances on wearable healthcare sensors based on paper materials in two perspectives, namely, instanta- neous monitoring of physical activities and rapid diagnosis of potential diseases using electrochemical detection methods.

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Physical monitoring

Continuous monitoring of physical signals and symptoms directly from human body using wearable devices is currently becoming a modern trend for fitness tracking and chronic disease prediction (e.g., osteoporosis precaution) in personalized health- care. For sensitive detection of human motions and preferable comfort in user experience, various soft materials have been adopted for prototyping wearable sensors in recent years [12,184,185]. Amid all the options, the ultra-thin and extremely light paper is generating increasingly extraordinary interest

because of the potential of inducing little irritation discomfort when directly attached onto the human skin. Meanwhile, the economical feature of paper plays a part in dramatically dropping the manufacturing cost of wearable monitoring sensors, which is of great significance for their widespread use in some developing countries and areas.

Recent studies have undertaken a number of fabrication techniques for depositing a sensing element on commercially available paper [27–29]. As an example, a high-resolution inkjet printing method has succeeded in fabricating an array of micro electro mechanical system (MEMS) sensors by patterning carbon ink onto chemically modified vellum paper [29]. The MEMS sensor array performed a sensitive response to cyclic upward-downward deflections [29], reflecting its promising application in body-worn pressure sensing and human motion monitoring. To further reduce cost, time and complexity of the fabrication process, a pen-on- paper paradigm [186] has emerged and subsequently produced significant inspiration for wearable electronics production. The utilization of omnipresent writing instruments unlocks the gate for consumers to doodle and write “on-demand” sensing components for unique “on-site” monitoring tasks [27,28]. A case in point is handling a pencil to write conductive graphite traces on a normal printing paper to realize the strain sensing function (Fig. 8a) [27]. Graphitic flakes rubbed off from the pencil rod

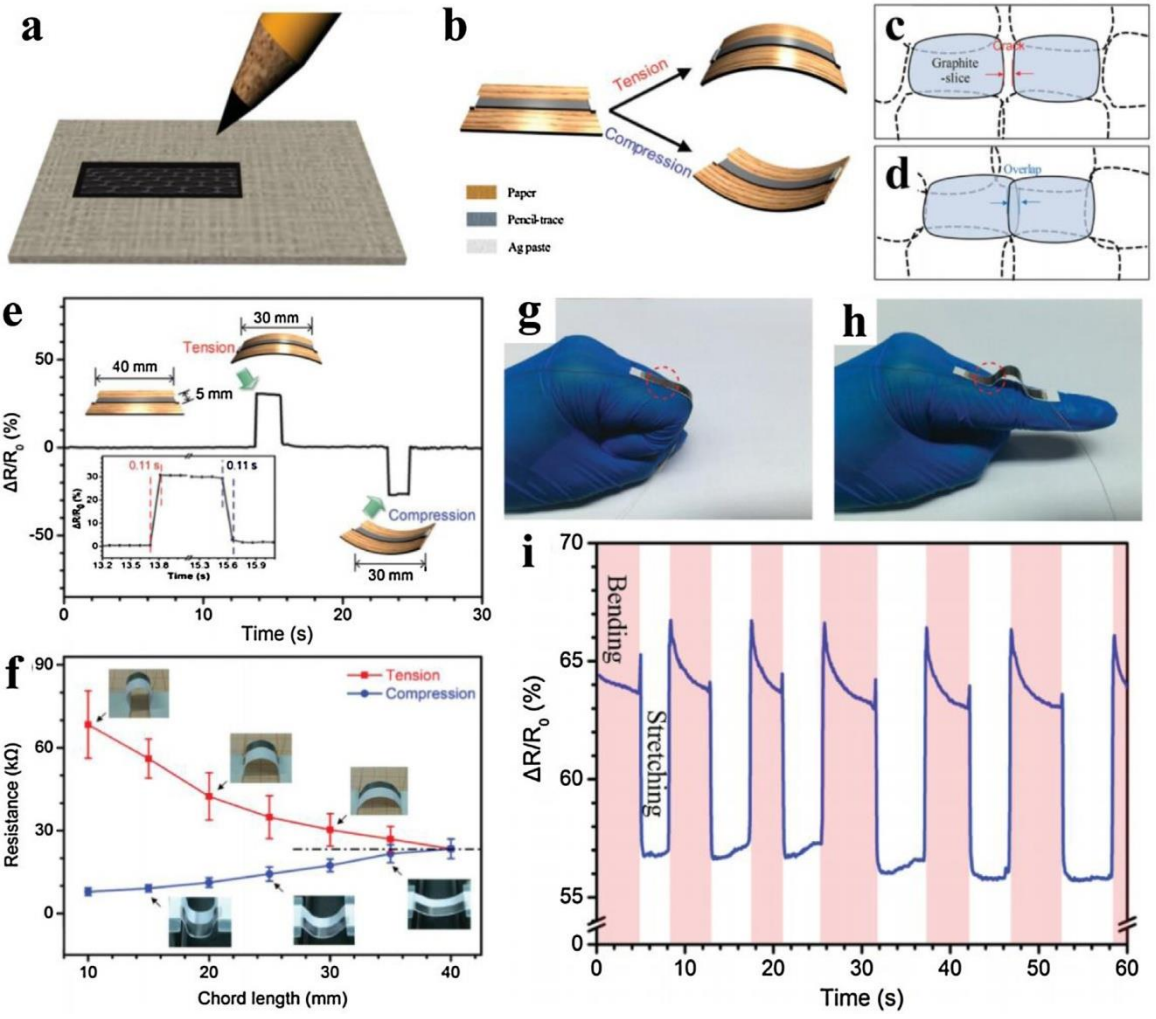

Fig. 8. Promising wearable applications as physical monitoring sensors. (a) Schematic of writing conductive traces on paper using a pencil. (b–d) Schematic diagram of the sensing principle under tension (c) and compression (d). (e) Response of the paper strain sensor with a response time of 0.11 s (inset). (f) Resistance of the paper sensor as a function of chord length. Inset photographs show the sensor in diverse tensile and compressive states. (g–i) Function demonstration of the strain sensor directly incorporated on finger in bending (g) and stretching (h) cases, and the normalized resistance change as a result of the sensing function (i) [27].

tended to penetrate into the porous cellulose structure of paperand form a uniform surface geometry, resulting in a significantlyenhanced conductivity of the pristine-insulating paper. The underlying mechanism of this paper-based strain sensor can be depicted as reversible microstructural changes of graphitic flakes subjected to tensile and compressive working conditions (Fig. 8b–d). As a result, the strain sensor showed an instant response(110 ms) (Fig. 8e) and exhibited a noticeably opposite resistancechange behavior under compression

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and tension (*i.e.*, resistancefell when compressed and rose when tensed) (Fig. 8f), which canbe used to speculate the bending direction of human motion. Themechanical fatigue test results suggested that this paper sensorremained stable and tenable under as many as 10,000 bending/ releasing cycles. Further, it was explored for on-finger motion detection and remarkable resistance variances under multiple deforming conditions of human skin (Fig. 8g–i) can be obtained.
Moreover, with the increasing focus on and rapid development
of graphene, the piezo-resistive effect of thin paper comprising graphene woven fabrics has also been investigated for wearable sensing with sensitivity and reliability [187]. Such graphene paper has a large gauge factor [188], making it easier to present observable resistive change as a detectable response for a small deformation (0.2%) of tiny physical motions, such as breathing, pulse, vocalization and facial expression changes [187].

Electrochemical detection
Point-of-care (POC) diagnosis featuring easy accessibility, portability, affordability and rapid operation, has recently been nominated as an increasingly important area of research that significantly contributes to the individual and public health community devoted to disease early screening, drug testing and environmental quality monitoring [189–192]. Amid many routes for getting access to POC diagnostics, the electrochemical detection approach has advantageous characteristics over other approaches, involving high detection sensitivity and low sensing limit [193– 195]. With the emergence of wearable technology in the past decade, pioneering scientific research has followed the pace of this trend, and the electrochemical sensor array can be directly worn onhuman body for health state monitoring by sampling sweat and sensing its chemical composition [4]. Furthermore, this combina- tion paved the way for massive adoption of paper, the dominant substrate material of electrochemical POC testing devices [196– 198], for wearable electrochemical sensing patches.

Physiological sensing. To date, multifarious paper-based electrochemical devices have achieved significant progress in human healthcare enterprise, mainly reflected by biochemical index evaluation and disease diagnosis at an early stage by electrochemically detecting antigens/antibodies [199–203], glucose [204–207], cholesterol [205], dopamine [208], adenosine [209] and ions [210,211]. In an electrochemical vertical-flow device fabricated by simple and effortless paper- origami (Fig. 9a,b), glucose monitoring function can be accurately realized by analyzing the corresponding potentiostatic coulometrydata [212]. This electrochemical testing system performs a highly sensitive and accurate detection of glucose amount with a maximum error of only 19% (Fig. 9c), unveiling its stunning perspective for promising wearable healthcare uses [212].
Electrochemical detections of analytes is normally performed in
a three-electrode system (*i.e.,* working electrode, reference electrode and counter electrode), in which the qualitative and/or quantitative information about the analytes can be obtained by recording the electrochemical signals (*e.g.,* current, potential or electric impedance) from the electrochemical reactions of analytes that occur on the working electrode surface. Thus, the composition, geometry and surface property of working electrode directly affects the performance of electrochemical detection. A significant number of efforts have been motivated to develop working electrodes with multiple geometries, compositions and enhanced electrochemical properties [213–215]. An example is the use of gold microwires and carbon microfibers to fabricate working electrodes. The prepared electrodes exhibited a higher mass transfer rate and lower capacitance compared with the working electrodes made from traditional millimeter scale gold and carbon

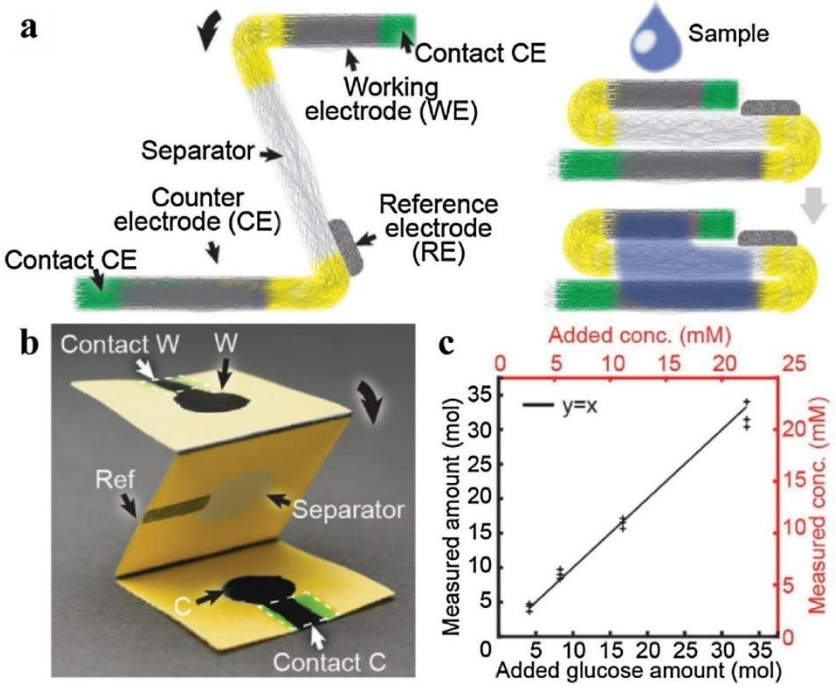

Fig. 9. Promising wearable applications as glucose sensing devices. (a) Side-view schematic of the paper-based electrochemical sensing device. (b) Digital photograph of the sensor before being folded into a stacked construct. (c) Potentiostatic coulometric results for glucose detection [212].

materials [215]. In addition, the working electrodes comprised of gold micromeshes were also prepared, in which the micromesh structure with increased surface area can provide sufficient sites for immobilization of some biological probes such as antibodies and nucleic acids, which is favorable for POC detection [215]. To further achieve the POC diagnosis using the electrochemical detection method in an inexpensive, simple and fast mode, additional attention has been paid to discovering novel manufacturing techniques for electrode prototyping [168,169,204,216,217]. Among these approaches, a writing scenariothat adopts a ubiquitous ball pen is the most prominent. For electrochemical purposes, the pen cartridge is refilled with conductive silver or carbon inks and hence the triple-electrode system can be directly doodled on paper with easy accessibility andrapid prototyping for glucose monitoring [204].

Gas sensing. Except for on-site evaluation of biomedical parameters, paper-based electrochemical devices have undergone implementation in continuous air quality and safety monitoring. By wearing these devices, users can be supplied with insight of ambient environment and be informed about health hazards that are hidden in air [218–220]. Using chemically modified CNTs and graphene as raw materials for electrode fabrication, on-paper sensors showed sensitive and specific testing results conducted on hazardous gases, such as ammonia, acetonitrile, toluene, cyclohexanone, as well as nitrogen dioxide, which can save human beings from these hazardous gases [219,220].
Further research interests have been stimulated on electro- chemical sensing of human respiration with a single piece of paper [221]. The operating mechanism of these paper sensors is based upon the fact that cellulose fibers in paper tend to absorb moisture from the ambient environment, and accordingly, the ionic conductivity of paper will increase/decrease due to the change of water content in cellulose (Fig.10a). Thus, the humidity variationinduced by cycles of expiration and inspiration can be converted into electrical signals and be electrically detected on the paper- based respiration sensors. Human breathing activity can be
seamlessly tracked with such sensors, as elucidated by thecapability to distinguish different respiration patterns (Fig. 10b, c). Moreover, the paper sensors can be incorporated into a commonfacemask, detecting respiration status in a wearable manner and transmitting signals to the customer's mobile device (*e.g.,* phone and tablet) for data storage and analysis and information display (Fig. 10d,e).
Electronic muscular actuators
Soft electronic muscular actuators [222–224] with superb performance and human friendly features are resulting in significant research interests in the new era of wearable consumer electronics because haptic and tactile sensing functionalities they supply to users bring us a step closer to burgeoning territories including bio-inspired robotics, human-machine interaction, virtual reality headsets and body-worn exoskeletons. Desirable actuators for these application perspectives are required to be compliant with human motion and be biocompatible. Therefore, soft and thin paper based on bio-origin cellulose or graphene has been developed as a category of potential substrate materials for electro-active artificial muscles [56,57,70,225,226].
Thin film of bacterial cellulose (BC) with superior biocompati-
bility was recently manifested to be feasible in producing soft electronic muscular actuators [225]. In this work, the 2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPO)-meditated oxidized func- tionalization process was first conducted on BC which promotes the formation of a facile membrane. After that, the obtained TEMPO-oxidized BC (TOBC) underwent sequential procedures of graphene loading and ionic liquid embedding for higher mechani- cal stiffness and ion exchange capacity enhancement, respectively. Then, with an extra coating of a conductive polymer, poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS), onboth the top and bottom sides of the composite film, a biofriendly graphene-reinforced, ionic liquid containing TOBC (TOBC-IL-G) paper-based electro-active artificial muscle was achieved. The surface morphology presented a fine dispersion of the TOBC-IL-G

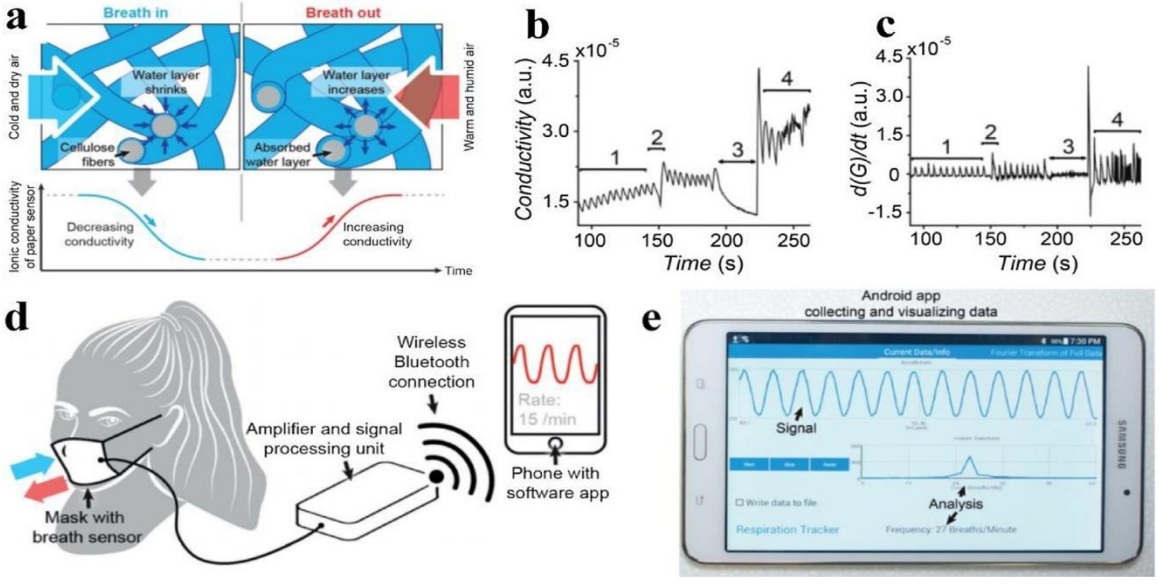

Fig. 10. Promising wearable applications as respiration sensors. (a) Operating mechanism of the paper-based respiration sensor. (b, c) Raw data (b) and high-pass filtered data (c) of the respiration signal recorded with four different breathing patterns: 1. normal breathing, 2. deep breath, 3. pause and 4. random breathing. (d) The entire respiration monitoring system consisting of a facemask embedded with a paper respiration sensor, a signal processing circuit and a mobile device for data display and analysis. (e) Digital photo of a tablet displaying the report of a user's respiration condition [221].

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composite induced by the strong and effective electrostatic interaction and hydrogen bonding between TOBC nanofibers, graphene and ionic liquid. Furthermore, the tensile test results of the TOBC-IL-G paper revealed a dramatic mechanical enhance- ment (23% greater tensile modulus and 75% higher tensile strength) compared with the pristine TOBC, which can be ascribed to the reinforcement of graphene. The enhanced electro-mechani- cal property due to graphene addition also results in superior actuation performance, including greater bending actuation and improved response time of the fabricated actuator compared with that made of pure TOBC (Fig. 11a,b). Further, a demonstration of this artificial muscular actuator was carried out to indicate its outstanding deformability under electro stimuli (Fig. 11c). In a most recent work, the hygro-expansive characteristic of paper was exploited to design and fabricate electrically triggered actuators on a thin sheet of paper/PEDOT:PSS composite [227]. Due to the fine embedding of the actuating element (*i.e.,* PEDOT:PSS) within the porous architecture of paper, these paper actuators operated evenly when bent and scratched.

Apart from cellulose, crumpled graphene paper with tailored
hierarchical surface structures have also been attempted in the application area of electro-triggered artificial muscle actuators [70]. The underlying mechanism for such tactic is the deformation of the graphene-elastomer laminate with thickness reduction and surface area increment of more than 100% prompted by the Maxwell stress [228], which occurs under direct loading of current voltage between a slice of graphene paper.

Memory devices

Paper has long been a predominant material for information containing and message delivery ever since its invention. In other words, paper is a classical and practical memory function medium. In recent years, consumer electronics has been experiencing an unprecedented trend of being bendable or foldable such as flexible displays [136,229,230] for televisions and wearable gadgets. For these smart computational systems, a memory device is undoubt- edly a fundamental component and plays a core role in media
recording and data storage. At this point, paper has again come out as an inexpensive, lightweight, green substrate candidate for fulfilling the concept of prototyping electronic memory with ultra- low cost and minimal motion impact on human daily activities when used in wearable systems.

One big challenge for fabricating paper-based electronic memory devices lies in the contradiction between the requirement of thin, uniformly deposited layers for typical memory devices and the micro-scale rough and porous structure of cellulose paper. To resolve this, a simple structured resistive random access memory was adopted to address the paper surface variations [137]. In such paradigm, a simple working structure of conductor-insulator- metal was configured by sequentially printing carbon paste, TiO_2 nanoparticles and silver nanoparticles onto paper surface with controllable resolution and great uniformity (Fig. 12a). The paper memory device behaved with well-defined ON/OFF switching ability (Fig. 12b), and the tunable memory window was realized by modulating the deposition thickness of the semiconducting TiO_2 nanoparticles (Fig. 12c). Note that such fabricated paper memory was mechanically robust enough to follow the motion of living objects once attached to their surfaces (Fig. 13a), thus paving the way for use in body-worn devices. To demonstrate this, an on-skin bending test was conducted. The obtained results indicated that the paper memory labels affixed on human body remained stable during 1000 switching cycles (Fig. 13b) and performed normally under stretching (Fig. 13c) and compression (Fig. 13d) of skin. Moreover, the paper memory device was reported to realize data elimination simply by burning or shredding. Thus, sensitive information can be secured from unexpected disclosure and leakage. In another work, a flexible memory device was demonstrated with nonvolatile memory effect of as high as six orders of switching resistance ratio under the utilization of nanofibrillated cellulose instead of commercial printing paper [231].

In addition, paper memory devices are also compatible with
other electronic components for function extensions in wearable electronics. For example, they can be integrated with radio frequency identification paper tags [232,233] for portable ticketing

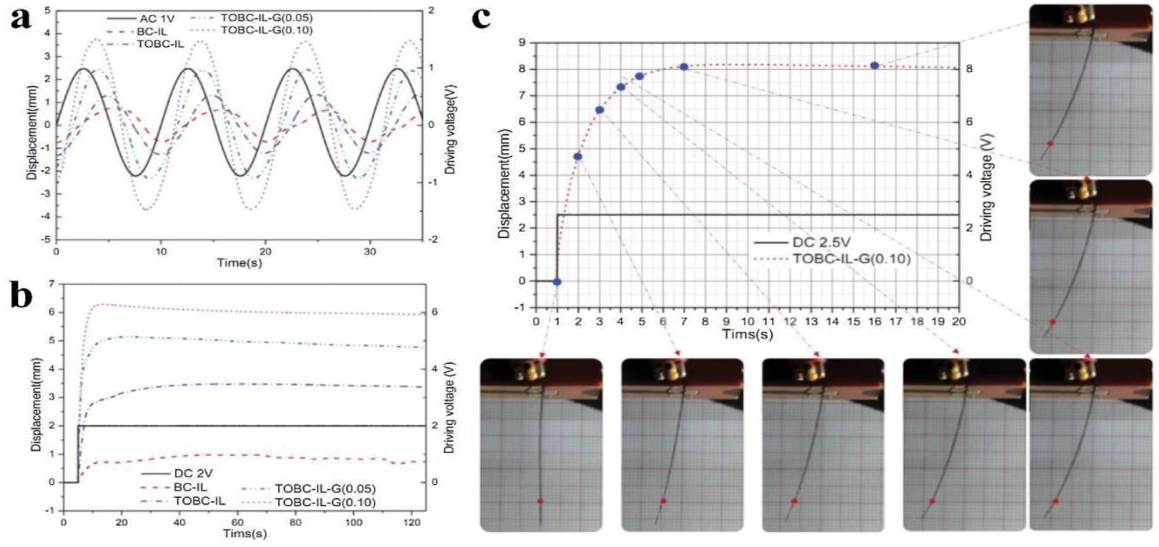

Fig. 11. Promising wearable applications as electronic muscular actuators. (a, b) Actuation responses under sinusoidal (a) and direct current (b) stimuli. (c) Mechanical deformation of the muscular actuator under direct current excitation. Insets: digital photographs of the electronic actuator with different bending extents as a time response [225].

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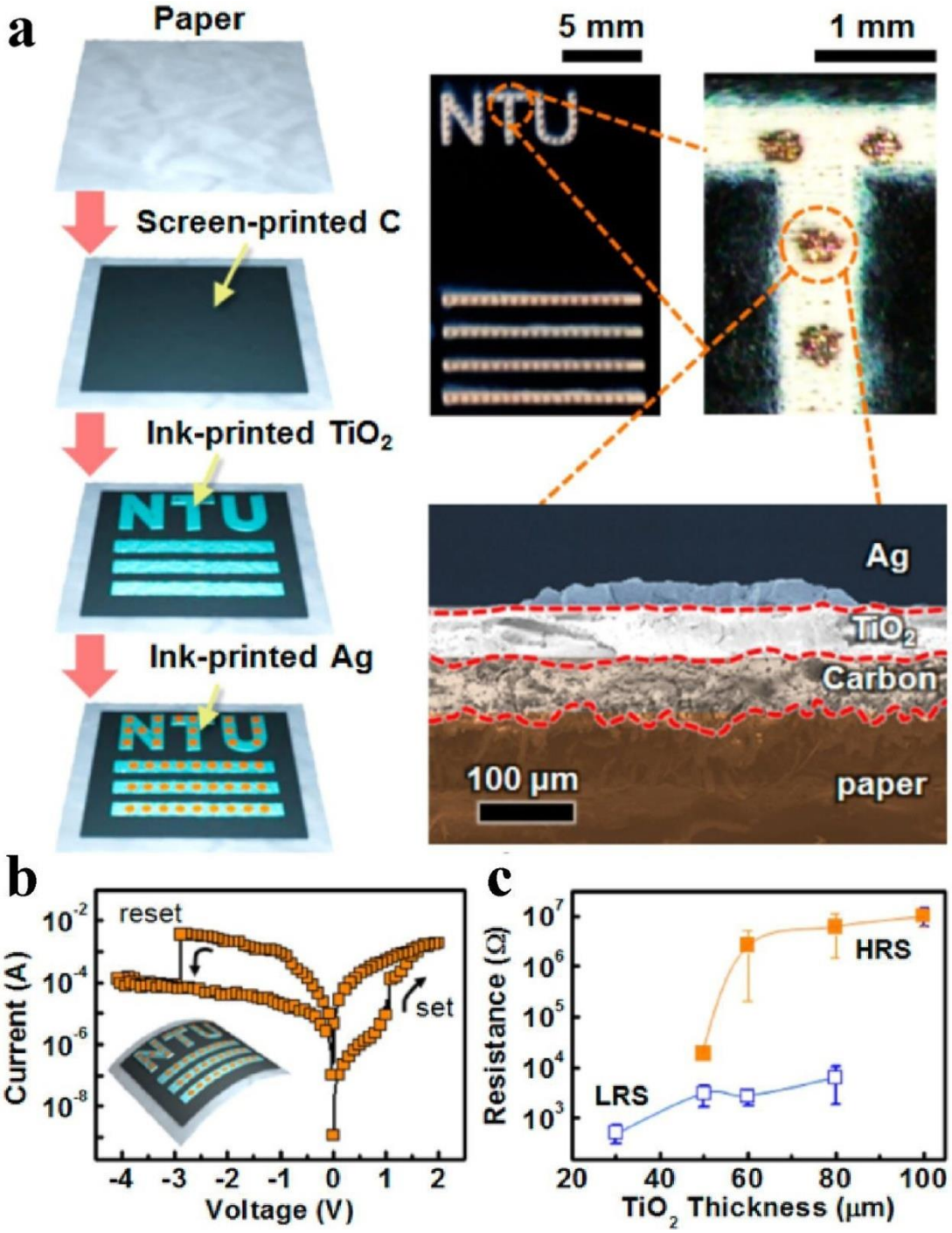

Fig. 12. Promising wearable applications as memory devices: fabrication. (a) Schematic diagram of the fabrication process. (b) Current-voltage characteristics of the paper memory. Inset is an optical image of the device. (c) Memory window at diverse TiO_2 layer thicknesses [137].

and security identification. Additionally, the paper-sustained antennas [44,186] can be used with memory devices aiming at wearable wireless data communications.

Energy storage/supply

Lightweight, inexpensive and bio-benign energy storage/supply devices are eagerly required in the upcoming decades to address the mounting energy demand issue of the increasingly fashionable wearable electronic functional devices and integrated systems [234–236]. This demand is promised to be filled along with the

exciting advances in paper electronics, because paper-based energy supply with appealing virtues of being thin, cheap and human friendly

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is exhibiting magnificent potential to provide power for paper electronic systems, and thus further fulfills the concept of self-powered, fully paper-based electronics [237–240].

Lithium ion batteries

In the past decades, as the most celebrated representative of therechargeable battery category, lithium ion batteries (LIBs) featur- ing high energy density and salient charging efficiency became popular for powering consumer electronic devices and green

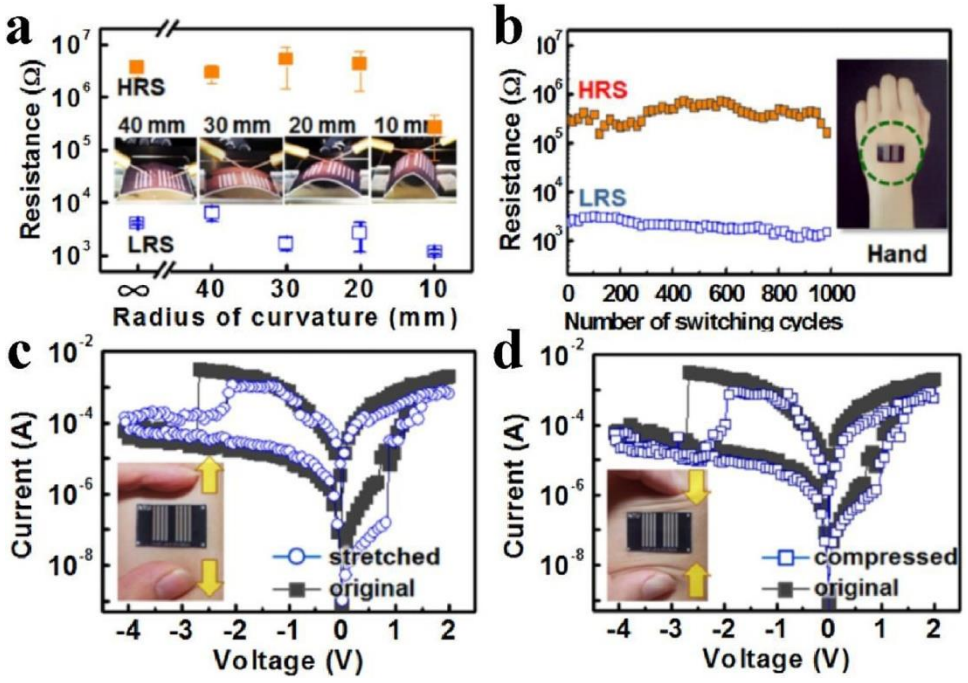
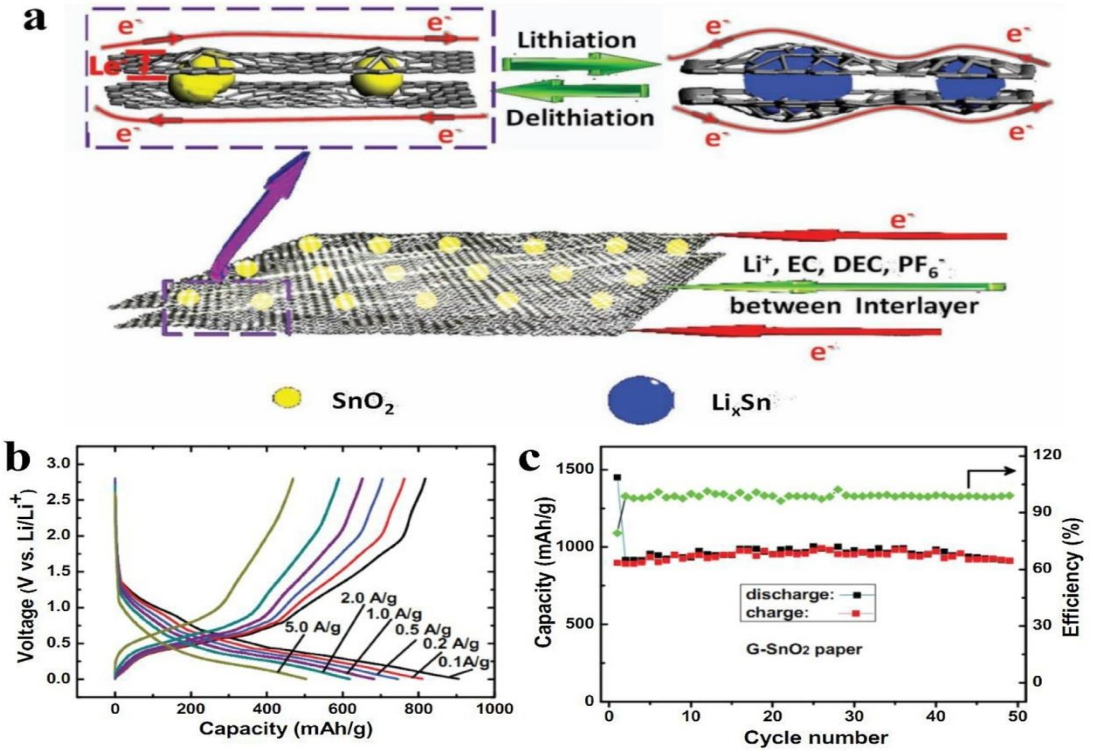


Fig. 13. Promising wearable applications as memory devices: characteristics. (a) Memory window of the as-fabricated paper memory device under mechanical deformations.



(b–d) Switching properties of on-skin memory under 1000 switching cycles (b), and mechanical deformation of stretching (c) and compression (d) [137].

Fig. 14. Promising wearable applications as energy storage/supply: lithium ion batteries. (a) Schematic demonstration of transportation passages for charge carriers provided with the unique sandwich structure of nitrogen-doped graphene/SnO_2 composite paper. (b) Charging/discharging characteristics and cycling stability of LIBs fabricated with the nitrogen-doped graphene/SnO_2 paper [249].

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electric automobiles [241–243]. Rhyming with the latest preva- lence of next-generation wearable electronics, efforts on defining the LIB with a revolutionary soft format and integrating it with body-worn, functional electronic gadgets for energy supply have been triggered in the material community [244–246]. Intriguingly, recent research advances have unveiled the feasibility of introduc- ing thin, lightweight and soft paper materials as backbone for LIBs, coupling with cutting-edge fabrication techniques to store and supply energy to the epoch-making wearable electronics with a slice of paper [247,248].

An LIB is generally comprised of two electrodes and a separator, in which charge-loaded lithium ions pass via electrolytes between cathode and anode to drive the circuit. Therefore, the selection of proper electrode materials is vital for the performance of LIBs. Supported by current scientific progress, thin papers made of electrically conductive carbonaceous derivatives (*e.g.*, graphene, carbon fiber and CNTs) have stimulated extensive interests in exploring their potential as electrodes for LIBs [58,248–251]. For example, a single piece of commercial paper was endowed with LIB functionality simply by laminating CNT films onto it as current collectors for electrodes, performing at an acceptable energy density of 108 mW H g^{-1} with a total battery thickness of only 300 μm [251]. To enhance the applicability of LIBs for high power situations, holey rGO papers with modified porosity distribution were used to fabricate electrodes, affording more and denser paths for transportation and storage of charge, and accordingly reinforcing the LIB capability at high charging/discharging rates [58]. Pursuing better battery performance for practical uses, thin membrane of carbon composite has also experienced applications for LIB electrodes. In a classical case, nitrogen-doped graphene/ SnO_2 hybrid papers were employed as anode for LIBs [249]. Notably, the composite was of a unique sandwich architecture with SnO_2 nanoparticles embedded between adjacent graphene layers (Fig. 14a), which was reported as a consequence of the presence of nitrogen atoms, disordering carbon construct and shortening transport path for charge carriers: lithium ions and electrons. Therefore, the enriched electrode electrical conductivity and battery rate capability were realized, with a specific capacity of 504 mW H g^{-1} at a current density as high as 5000 mA g^{-1} (Fig. 14b). In addition, this distinct structure contributed to satisfactory cycling performance (Fig. 14c) by rendering elasto- meric buffer space for accommodating SnO_2 volume variance when lithium ion was inserted or extracted.

In addition to electrodes, separator in LIBs is important for realizing the function of electric power storage and supply. It is a permeable film that is between a cathode and anode and that isolates them in case of short circuits while also provides transport passages for ions. Taking this functionality into consideration, paper materials with a porous internal structure that can offer sufficient transportation paths for ion charge carriers have a significant potential to be employed as separators. This potential is being gradually confirmed with the evidence that paper has been successfully equipped in LIBs as a separating component, furnish- ing mechanical support and electrochemical functionality [38,119,251].

Supercapacitors

Due to the outstanding advantages of higher energy density and shorter charging period over its counterparts, a supercapacitor with an electrochemical double layer is currently attracting more research attention than ever with unparalleled potential for serving paper-based energy solutions [125,132,252–254]. In a classic strategy, a pencil was adopted to directly write conductive graphitic patterns on a thin sheet of Xerox paper to create a classical supercapacitor structure, with graphitic traces as electro- des and paper as a separator, in a simple and ultra-economical manner [253]. A sophisticated and easily scalable "soak-and- polymerization" technique has also been utilized for the large scale manufacturing of wearable power supplies, with a promoted areal capacitance (0.42 F Cm^{-2}) and high energy density (as much as 1 mW H Cm^{-3}) by coating a highly conductive polypyrrole (PPy) on commercial printing paper [125]. However, there is a general

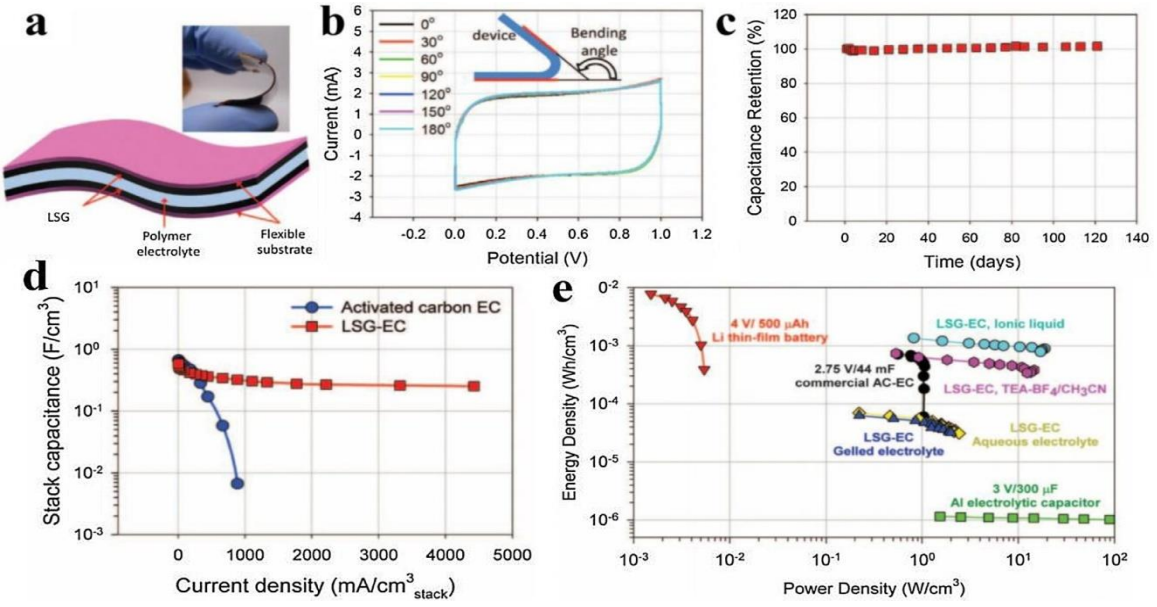

Fig. 15. Promising wearable applications as energy storage/supply: supercapacitors. (a) Schematic and digital photo of the supercapacitor based on graphene paper. (b) Bending effect on the current-voltage characteristics of the supercapacitor. (c) Long-term durability of the graphene-based supercapacitor. (d) Stack capacitance as a function of current density depicting the high rate performance of the supercapacitor. (e) Energy and power densities of the fabricated graphene supercapacitor compared with its counterparts [259].

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challenge for such energy supply devices based on conductive paper that can be depicted as the dramatic capacity decline under high charge/discharge rates [239]. To resolve this, an approach based on carbon filament reinforcement was proposed [132]. Specifically, chopped carbon filaments were blended with a paper- thin composite of PPy and cellulose, which resulted in enhanced electrochemical performance of the composite paper by decreas- ing its self-resistance and contact resistance [132]. Accordingly, the as-fabricated paper-based power source presented an attainable performance at charge/discharge rates as high as 500 mV s^{-1} [132], representing promising perspectives for energy supplies in high- power working conditions.

The intrinsically superb electrochemical property has endowed carbon-based materials with incomparable potential for energy storage/supply [58,255–257]. Graphene paper, a typical represen- tative of the novel carbon materials family, has gained most favor from energy solution makers for its excellent electrical conductiv- ity (55 S m^{-1} [258]), high surface area (1.52 m^2 mg^{-1} [259]) and robust mechanical stability. These unique properties benefit graphene paper to behave as both an active electrode and a current collector, and hence simplify the manufacturing process by excluding the additions of active materials and metal for collecting current, which are commonly required for the stereotyped fabrication. In one case, thin films of graphene were initially attained via reduction of GO irradiated by an infrared laser from a CD optical drive [259]. Next, two slices of the obtained graphene product were readily pasted onto both sides of a separator membrane to finish the easy-accessible and all-solid-state fabrication of an ultrathin (<100 μm) supercapacitor (Fig. 15a) with committed performance facing severe mechanical deforma- tion (Fig. 15b), over four-month idle period (Fig. 15c), superb rate performance (Fig. 15d), intensive energy density of up to 1.36 mW H Cm^{-3} and power density as high as approximately 20 W Cm^{-3} (Fig. 15e) [259].

Others

Regarding the burgeoning field of functional wearable elec- tronics, energy supply that can be incorporated in garments or attached to human body, harvest energy from human motion and transform it into electricity to power the entire electronic system [260,261] is needed. It has been recently reported that commer- cially available printing paper has the capability to serve as the base material for realizing energy generator with elaborate coating of silver and polytetrafluoroethylene on paper and careful assembly of the fabricated composite sheets [123]. The paper generator works under the principle of electrostatic effect and appears to efficiently harvest energy from tiny mechanical deformations (Fig. 16) [123].

Moreover, other power sources, such as solar cells [262,263] and fuel cells [264,265], have been realized with paper materials, strengthening the family of paper-based energy storage/supply and making a great step forward towards their practical applica- tion.

Conclusions and perspectives

Dislodging constraints of stiff profiles and rigid surfaces out of stereotyped electronic devices, a burgeoning and revolutionary form of electronics that can operate under mechanical deforma- tion with virtually no deterioration in performance is provoking innovations in designing smart electronic gadgets and accessories that can seamlessly fit in garments or even be directly affixed to human bodies with comfort and conformance, namely, wearable electronics. Selection of proper substrate materials with a unique mechanical deformability is expected to be a vital step towards making electronics conform to and be worn on the body. The diversification of soft materials provides a wide range of options for substrate uses. Among the potential candidates, paper materials (*e.g.,* cellulose paper, graphene paper, carbon micro-

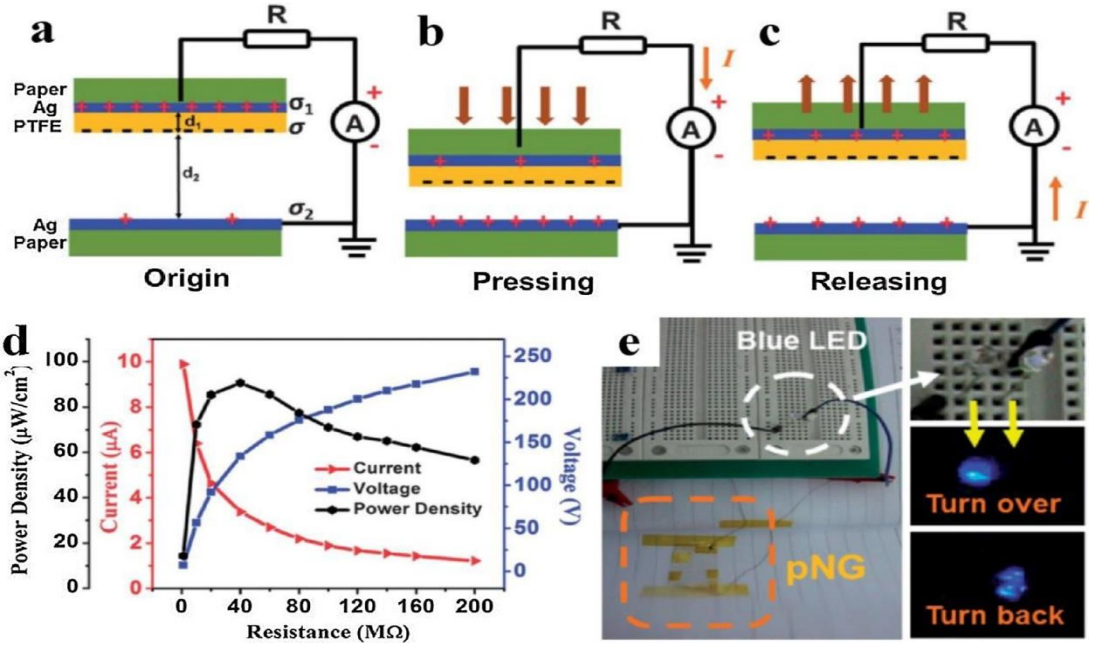

Fig. 16. Promising wearable applications as energy storage/supply: nanogenerator. (a–c) The working principle of the paper-based nanogenerator with external loading under original (a), pressing (b) and releasing (c) conditions. (d) Power density, current and voltage of the output as a function of the loading resistance. (e) Optical image to demonstrate electricity harvesting from mechanical deformation [123].

fiber paper, carbon nanotube paper, composite paper, *etc.*) are captivating a spectacular surge of research interest that can be ascribed to light weight, low manufacturing cost, mechanical deformability and stability, and bio-friendly and non-toxic quality that these materials have in common. As for the electrical properties, the paper based on carbon derivatives is pristinely conductive, and cellulose-based paper can benefit from its interior porous architecture in electrical conductivity reinforcement despite its inherent insulating property. Along with ingenious fabricating technologies (*e.g.*, coating, printing, writing and microfluidics strategy), these superior characteristics have trig-gered the use of paper for electronics production in potential application sectors of on-site physiological monitoring for health- care evaluation and diagnostics, wearable electro-active artificial muscles for human-machine interaction and disability rehabilita- tion, clothes-integrated memory storage for identification and body-worn energy supply for the whole wearable electronic systems.

The growing demand of wearable electronic products for personal healthcare, entertainment, industrial and military pur- poses are stimulating the rapid industrialization of recently uprising research achievements. However, some challenging issues are suggested to be approached or circumvented before the fabricated paper electronics can step into the market for practical uses. First limitation lies in the fact that the current approaches for manufacturing paper electronics either require professional and bulky equipment (such as coating and printing) or suffer from the disability to scale up for mass production (such as writing and microfluidic approaches). Tedious post-treatment procedures with extreme conditions including high temperature, long period and vacuum ambiance are required in some certain cases, which further increases the financial cost and energy consumption. It is also worth noting that when taken into real-life applications, the electronic devices made of paper may not be as robust as tested in the research lab, accounting for potential chemical corrosion of moisture and mechanical overloading on brittle layers, which can cause performance failure and further curtail the lifetime of such electronics. The final question is about the integrity of paper electronics. Multiple electronic functionalities including sensing, actuation and power supply have been successfully realized on paper substrates, but fully integrated paper-based circuits that are critically vital for electronics are yet to be released, and when they will be invented still remains uncertain.

Thus, there exists an urgent demand for more acquisitions from related sciences and studies to consolidate and promote the developing trend of paper-based wearable electronics in the following years. To begin with, because it is commonly accepted that a strong bond exists between material structures and physical/ chemical properties, researchers in material science are firmly encouraged to spare no efforts in completing knowledge of currently observed paper material constructs, developing novel paper structures with unique features, and most importantly, integrating some of the captivating properties (*e.g.*, optical transparency, magnetic feature, acoustic property and heat transfer) into paper-based wearable electronic devices and gadgets for improved functionality and robustness. Next, objective is of significant strategic value, which is a breakthrough in establishing tractable and scalable manufacturing techniques that can simul- taneously lower the barrier and increase the mass production. This is suggested to be shortly placed on the agenda. In addition, attention should be paid to introducing protective layers to encapsulate the paper base from external destruction. Without any doubt, these protective barriers should be thin, soft, transparent and most significantly, with no impact on the smooth operation of original paper electronics. Further outlook goes to the integration of functional elements with interconnects, energy supply and even

more advanced logic programmable units that are entirely fabricated on paper to actually fulfill the concept of paper electronics for practical uses as a miniaturized smart operator on the body. Meanwhile, the combination of a paper display will moreover prompt the integration and user experience in specific application perspectives. The goal of human physiological tracking will be greatly satisfied with the integration of a display that can help provide real time data of physical activity and valuable insight into the health condition of the user. To sum up, with all the upgrades and progress done and dusted, paper-based electronics will bring a revolutionary development to the wearable electronics industry in material selection and will everlastingly thrive for making the world a better place to live in.

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