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Application of Wearable Electronics on Paper Based Materials

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ABSTRACT

Skin-interfaced wearable hardware can track down an expansive range of uses in medical care, human-machine interface, mechanical technology, and others. The best in class wearable hardware ordinarily experience the ill effects of exorbitant and complex manufacture methods and nonbiodegradable polymer substrates. Paper, including trapped miniature or nano-scale cellulose filaments, is viable with versatile manufacture methods and arises as a manageable, reasonable, expendable, and biocompatible substrate for wearable hardware. Given different alluring properties (e.g., breathability, adaptability, biocompatibility, and biodegradability) and rich tunability of surface science and permeable designs, paper offers many invigorating freedoms for wearable gadgets. In this audit, we initially present the fascinating properties of paper-based wearable hardware and methodologies for cellulose changes to fulfill explicit requests. We then outline the uses of paper-based gadgets in biosensing, energy stockpiling and age, optoelectronics, delicate actua-pinnacles, and a few others. At last, we talk about certain moves that should be promotion dressed before viable utilizations and wide execution of paper-based wearable gadgets.

INTRODUCTION

Emerging wearable electronics have achieved significant advancements in a wide spectrum of applica- tions, spanning from healthcare and human-machine interface to soft robots and virtual and augmented reality (Bariya et al., 2018; Kim et al., 2019; Ray et al., 2019; Someya and Amagai, 2019; Someya et al., 2016; Yang and Gao, 2019). Existing wearable electronics usually rely on clean-room-based fabrication techniques and/or nonbiodegradable polymer supporting substrates (e.g., polyimide [PI], polyethylene terephthalate [PET], and silicone elastomers) (Liu et al., 2017; Sun et al., 2018; Xu et al., 2020a; Yu et al., 2019). Its commercial translation is therefore limited as a result of the high manufacturing cost. Moreover, next-generation wearable electronics should be skin friendly (e.g., breathable), disposable, and one-time use to minimize the risks of inflammation and infections. Considering the wide distribution and implemen- tation of wearable electronics in the future, the accumulation of disposed electronic wastes will require sub- stantial demand for the landfill space and cause unfavorable environmental issues. Such requirements contradict the current materials selections of wearable electronics as they are generally nonbreathable, nonbiodegradable, and expensive. Therefore, researchers are motivated to seek sustainable alternatives with desired features that can address the aforementioned handicaps.

Examples include packaging, display, and information storage. Owing to the hierarchically entangled cellulose fiber structures, paper features many intriguing properties in terms of rich surface chemistry (i.e., hydrophilic hydroxyl groups) and pore size tunability. Moreover, the superior tailorability of paper granted by kirigami and origami designs brings more possibilities for the development of paper-based three dimen- sional devices (Ding et al., 2016; Lin et al., 2017; Xu et al., 2020b). In addition, paper's intrinsically sustainable, breathable, flexible, biocompatible and biodegradable nature broadens its promising and versatile appli- cations in wearable electronics (Figure 1A). Leveraging scalable manufacturing approaches (e.g., inkjet printing, screen printing) or highly customizable and easily accessible writing process (e.g., pencil or pen writing), myriads of applications have been explored for paper-based wearable electronics, including dis- playing (Asadpoordarvish et al., 2015; Ha et al., 2018; Jeong et al., 2019), sensing (Mahadeva et al., 2015)

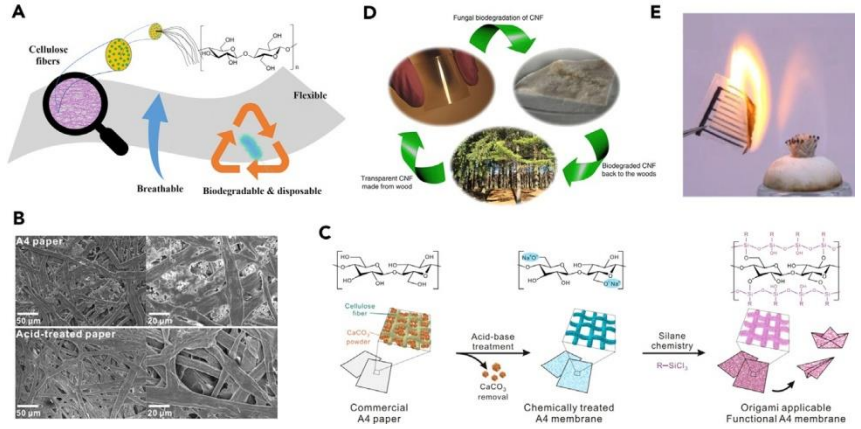

Figure 1. An overview of desirable merits of cellulose paper

(A) Schematic illustration of cellulose paper with high flexibility, breathability, biodegradability, and disposability.

(B) Scanning electron microscope (SEM) images of pristine and acid-treated commercial paper. Adapted with permission from Ahn et al. (2020). Copyright 2020, American Chemical Society.

(C) Acid-base treatment and surface modification of cellulose paper with trichlorooctylsilane. Adapted with permission from Ahn et al. (2020). Copyright 2020, American Chemical Society.

(D) A typical life cycle of the biodegradable, eco-friendly, and sustainable cellulose paper. Adapted with permission from Jung et al. (2015). Copyright 2015, Nature Publishing Group.

(E) Incineration of a paper-based electronic device. Adapted with permission from Gao et al. (2019a). Copyright 2019, American Chemical Society.

Tai et al., 2020), optoelectronics (Ha et al., 2018), energy harvesting and storage (Yao et al., 2017; Zhang et al., 2015), and many others (Hamedi et al., 2016; Liao et al., 2020).

In this review, we discuss the potential, progress, and challenges of cellulose paper-based wearable elec- tronics. Specifically, we first introduce the critical mechanical and chemical features of cellulose papers, which can meet the requirements of human- and eco-friendly wearable electronics. Next, we overview the recent research of paper-based electronic devices, including biosensing, energy harvesting and stor- age, soft actuators, optoelectronics, and several others. Finally, we discuss the opportunities and future challenges. We believe that paper-based wearable electronics can find a breath of potential applications in low-resource environments and home-based, personalized healthcare as a sustainable platform.

PROPERTY DEMAND

Wearable electronics require mechanically flexible and stretchable yet robust supporting substrates. At the molecular level, the inter- and intramolecular hydrogen bonds among densely packed hydroxyl groups, together with Van del Waals interactions, in cellulose paper generate strong interfacial interactions (Li et al., 2021). Moreover, randomly distributed fibrous networks provide physical entanglement that forms porous structures (Figure 1B), which further enhance the flexibility (Ahn et al., 2020). These features offer lightweight, flexible, yet mechanically tough properties. Commercial copy paper is typically filled with mineral fillers such as calcium carbonate ($CaCO_3$), chalk, and clays to improve light scattering, ink absorbency, and surface smooth-
ness (Tobjörk and Österbacka, 2011). Removing these fillers can tune the porosity, pore size distribution, and
mechanical strength. For example, acid and/or base treatments of a commercial A4 paper increase the pore size and porosity from 0.31 mm and 50.3% to 12.2 mm and 82.9% (Figures 1B and 1C) (Ahn et al., 2020). As a result, the mechanical toughness along with fracture strain decreased somewhat, whereas the tensile strength increased from ~20 to ~30 MPa. In addition, the pristine porous fibrous network provides excellent breath- ability (>6,000 g m^{-2} day^{-1}) (Xu et al., 2020b) that far surpasses that of human skin (204 g m^{-2} day^{-1}) (Chao et al., 2018), which is critical considering long-term and continuous skin-interfaced applications. Also, high stretchability can be achieved in paper with judiciously engineered kirigami designs.

The abundant hydroxyl groups of cellulose fibers are advantageous for microfluidics where liquid transpor- tation is driven by capillary force. The rich surface chemistry has also been explored when integrating paper with conductive materials (e.g., metallic nanomaterials, carbonaceous nanomaterials) (Zhang et al., 2018).

However, the intrinsic hygroscopic nature of cellulose paper inevitably compromises the stability and robustness because of unfavorable mechanical degradation and irreversible deformations in wet environ- ments (Tobjörk and Österbacka, 2011). This handicap can be resolved by surface chemistry engineering.
For example, silanization of paper results in substantially increased hydrophobicity (Figure 1C) (Ahn et al., 2020). Particularly, an omniphobic "R^F" paper was fabricated by vapor-phase silanization of paper with fluoroalkyl trichlorosilanes, while preserving its intrinsic porous structure (Glavan et al., 2014). This strategy enables a hydrophobic and oleophobic paper that repels both aqueous and organic solutions such as blood, significantly improving the stability and robustness in biological environments. Inkjet print- ing of conductive inks on the "R^F" paper also exhibits higher lateral printing resolution down to ~28 mm (Lessing et al., 2014). In addition, other device fabrication methods, which generally involve deposition of hydrophobic reagents on paper, are also demonstrated, including analog printing, wax printing, flexog- raphy printing, and screen printing (Mahadeva et al., 2015).

Given the potential of green and environmentally friendly wearable electronics, cellulose paper is playing increasingly critical role as a sustainable supporting substrate. While most polymers take hundreds of years to decompose in natural environments, cellulose paper can be decomposed within several weeks by a va- riety of microorganisms (fungi, bacteria, and yeasts) that exist naturally in soil (Coughlan, 1991). Figure 1D presents a typical life cycle of cellulose nanofibrils that are originally extracted from wood, subsequently degraded by fungi, and recycled back for forestry fertilizer (Jung et al., 2015). Specifically, two types of fungi, brown rot fungus *Postia placenta* and white rot fungus *Phanerochaete chrysosporium*, have demon- strated the capability of cellulose biodegradation. The disposability of paper-based electronics is also exemplified by a simple incineration process (Figure 1E) (Gao et al., 2019a). Moreover, cellulose is consid- ered as a biocompatible material and has been used in numerous bioengineering-related fields. In addition to skin-interfaced electronic devices that are covered in this review, other application areas of cellulose ma- terials include tissue engineering, drug delivery, medical microfluidic diagnostics, and wound healing (Cate et al., 2015; Czaja et al., 2006; Gong and Sinton, 2017; Hickey and Pelling, 2019; Sun et al., 2019). Owing to the aforementioned properties, cellulose paper emerges as an appealing platform for the fabri- cation of wearable electronics.

APPLICATIONS OF PAPER-BASED WEARABLE ELECTRONICS

Biosensing

Wearable electronics can provide continuous, long-term monitoring of dynamic changes of physiological signals and hold great promise in fitness tracking, medical diagnostics, and human-machine interface (Bar- iya et al., 2018; Kim et al., 2019; Ray et al., 2019; Someya and Amagai, 2019; Someya et al., 2016; Yang and Gao, 2019). Owing to the intrinsic nonconductive nature (10^{11}–10^{15} U sq^{-1} at a relative humidity of
20%–40%) of cellulose paper (Tobjörk and Österbacka, 2011), conductive and semiconductive materials
are required to make paper-based electronic devices. A number of solution printing techniques have there- fore been developed for the fabrication of paper-based wearable devices as they are typically fast, inex- pensive, and easily customizable. The existing techniques include inkjet printing (Choi et al., 2016; Huang et al., 2014; Lessing et al., 2014), screen printing (Adkins et al., 2017), spray coating (Asadpoordarvish et al., 2015), vacuum filtration (Li et al., 2014), pen writing (Liao et al., 2015, 2020; Russo et al., 2011; Xu et al., 2020b; Zheng et al., 2011), and dip coating (Ding et al., 2016; Gong et al., 2014). Also, ***in situ*** synthesis of functional materials by chemical reduction or polymerization is another alternative strategy (Zhang et al., 2018). Selection of printing techniques should be based on the demands for lateral resolution, printed thickness, homogeneity, printing speed, materials, and ink properties. Although optimization by mixing with mineral fillers has already been made in commercial copy paper to minimize surface roughness, chal- lenges still remain when printing nanomaterial-based inks. For example, ink absorption into the porous substrate typically occurs as a result of the capillary force from micro-sized pores (Choi et al., 2016). Further- more, nonuniform pore size distribution within the paper matrix causes random spreading of the ink drop- lets, together with its hygroscopic expansion, thereby leading to decreased lateral resolution (Choi et al., 2016; Lessing et al., 2014). Some methods have been proposed to alleviate this issue, such as surface sila- nization of paper with fluoroalkyl trichlorosilanes (Lessing et al., 2014) and surface coating with a primer layer where cellulose nanofibrils are deposited to form a well-developed nanoporous structure with mini- mized surface roughness (Choi et al., 2016).

During the past several years, the studies on paper-based wearable electronics have grown substantially, aiming to collect a variety of information such as temperature (Xu et al., 2020b), strain (mainly bending

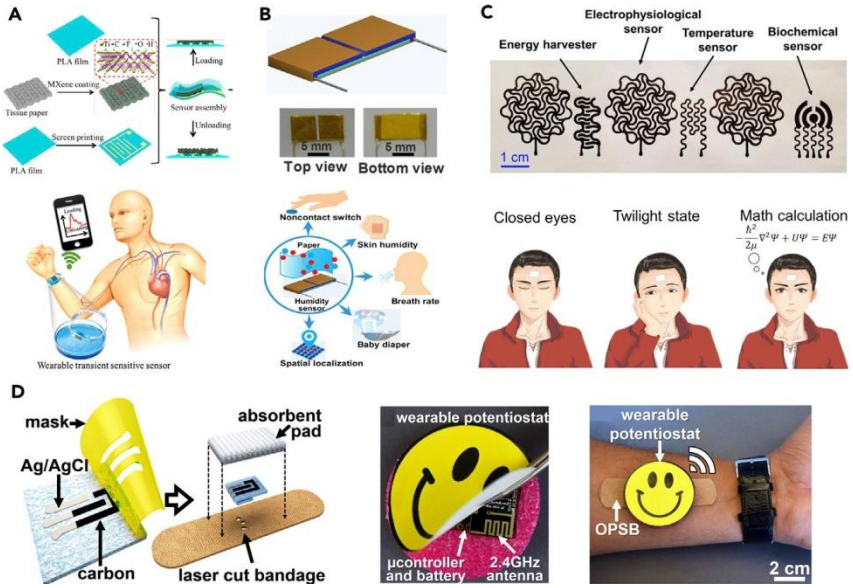

Figure 2. Paper-based wearable biosensors

(A) Paper-based wearable pressure sensor for pulse waveform measurement. Adapted with permission from Guo et al. (2019). Copyright 2019, American Chemical Society.

(B) Paper-based wearable humidity sensors for multiple biomedical applications. Adapted with permission from Duan et al. (2019). Copyright 2019, American Chemical Society.

(C) Pencil-paper-enabled multimodal wearable device and its application in monitoring of the human mental state. Adapted with permission from Xu et al. (2020b). Copyright 2020, National Academy of Sciences.

(D) Paper-based wearable smart bandage for electrochemical sensing. Adapted with permission from Pal et al. (2018). Copyright 2018, Elsevier.

strain) (Hua et al., 2016; Liao et al., 2015), pressure (Gao et al., 2019a; Gong et al., 2014; Guo et al., 2019; Zhong et al., 2015), light (Lin et al., 2017; Pataniya and Sumesh, 2020), biopotential (Sadri et al., 2018; Xu et al., 2020b), pH (Xu et al., 2020b), gas (Huang et al., 2014; Lin et al., 2014; Liu et al., 2014; Mirica et al., 2012, 2013), humidity and respiration (Duan et al., 2019; Güder et al., 2016), and biochemical compositions (Gong and Sinton, 2017; Pal et al., 2018). In these devices, paper can serve as either breathable and biocom- patible supporting substrates or active materials. Leveraging the piezoresistive characteristic of many func- tional materials, such as carbon-based materials, metals, metal oxides, and conductive polymers, numerous paper-based strain and pressure sensors have been developed (Mahadeva et al., 2015). The enabled devices can provide real-time and continuous monitoring of many vital signals, such as respiration, pulse waveform, and acoustic vibration (Figure 2A) (Güder et al., 2016; Tao et al., 2017). Leveraging its hy- groscopic nature, cellulose paper has been used to make humidity sensors for various applications in noncontact switch, skin humidity, breath rate, and baby diaper (Figure 2B) (Duan et al., 2019). Moreover, paper-based VOCs (volatile organic compounds) sensors are emerging as an inexpensive and versatile alternative of conventional bulky and expensive gas chromatography mass spectroscopy (GC-MS) tech- nique. The applications include medical diagnosis, environmental monitoring, and food quality assess- ment. For example, breath VOCs analysis has been employed for noninvasive diagnostic of a breadth of diseases including ovarian carcinoma, cancers, and end-stage renal disease (Broza et al., 2019). By modi- fying paper with various functional materials (e.g., carbon nanotubes, graphene, polypyrrole, and pencil leads), numerous paper-based chemiresistive VOCs sensors have been developed for analysis of ethanol, NO_2, and NH_3 (Mirica et al., 2012; Tai et al., 2020). The enabled paper-based VOCs sensors generally demonstrate good reproducibility and sensitivity (down to ~80 ppb) (Liu et al., 2014), which largely de- pends on the active sensing materials.

In addition to piezoresistive and chemiresistive sensors, biopotential sensors that monitor the electrical properties of biological tissues provide a wealth of valuable information. For example, electrocardiogram (ECG) represents the electrical activity of heart muscle and remains an essential role in cardiac assessments. To achieve high-fidelity recording of electrophysiological signals, an intimate contact between the sensing

electrodes and skin is required. Although paper is generally not sticky, silicone adhesive can be spray coated on its surface to resolve this issue (Xu et al., 2020b). The resulting paper substrate exhibited reliable and robust adhesion with skin while preserving its intrinsic breathability. In addition, there are various types of commercial adhesive cellulose papers (e.g., Aquasol adhesive water-soluble paper, Post-it Notes paper) that can meet this particular requirement without needing additional coating of silicone adhesive. Owing to low-cost resources, ease of fabrication, and abundant potential designs, paper-based wearable elec- tronics show great potentials in electrical and optical stimulation, temperature, ECG, EMG (electromyo- gram), and EEG (electroencephalogram) recordings (Figure 2C) (Broza et al., 2019; Xu et al., 2020b).

Given the rich surface chemistry, intrinsic hydrophilicity, and hierarchically porous structure, cellulose pa- per is particularly attractive for biochemical sensors. The hydroxyl groups together with micro-sized pores provide the capillary force that drives the transportation of biofluids, which provides inherent capability for microfluidic devices. By selective surface functionalization with hydrophobic moieties, microfluidic chan- nels can be constructed on paper (Mahadeva et al., 2015). Although continuous operation driven by the capillary force in a long-term manner has been rarely demonstrated by paper-based microfluidic devices, the physiologically relevant pressure from sweat glands (~2 kPa) may enable continuous sweat transporta- tion as demonstrated by elastomer-based microfluidic devices (Reeder et al., 2019; Yang et al., 2020; Yu et al., 2020). Although colorimetric detection is widely studied, electrochemical sensing is highly desirable owing to its quantitative analysis and insensitivity to light, dust, and insoluble compounds. This technique is extensively explored for a number of analytes such as glucose, cholesterol, drugs, pH, uric acid, potassium ferricyanide, L-lactate, and alcohol in blood, urine, and sweat samples (Nie et al., 2010; Pal et al., 2017, 2018; Xu et al., 2020b). For example, a paper-based wearable omniphobic smart bandage (OPSB) has been developed for the detection of pH, uric acid, and potassium ferricyanide in human open wounds, where carbon and silver/silver chloride inks are screen printed as working, counter, and reference electrodes, respectively (Figure 2D) (Pal et al., 2018). Subsequent interfacing the OPSB with a wearable potentiostat, battery, and antenna completed the device fabrication, enabling a wearable system for the early detection of pressure ulcers (Figure 2D). Paper-based point-of-care diagnostic devices are well studied in terms of the sample collection and analysis. Such biosensing applications can be exemplified by modifying paper electrodes with ion-selective membranes for potentiometric ion sensors. Various ion-selective electrodes are adopted for the detections of Cd^{2+}, Ag^{+}, K^{+}, NH_4^{+}, Cl^{-}, and Na^{+} (Hu et al., 2016; Mensah et al., 2014; Novell et al., 2012; Sakata et al., 2020). Given the intrinsic high porosity of cellulose paper substrate, these devices usually possess enhanced active surface area and therefore exhibit higher sensitivity. Of note, direct pencil drawing has also been demonstrated for the fabrication of wearable sweat sensors (Xu et al., 2020b). Compared with other often-used microfluidic devices (Bandodkar et al., 2019; Reeder et al., 2019; Yang et al., 2020), cellulose paper-based microfluidic devices exhibit distinct features, including (1) flexibility and breathability; (2) disposability; (3) inexpensive and scalable manufacturing pro- cess. However, challenges remain as encapsulation is required to minimize sample contamination and cross talk of different biosensors. Besides, the reproducibility and complicated calibration procedures are still main issues of paper-based biochemical sensors that need to be addressed by optimizations and innovations in the manufacturing process.

As compared with traditional wearable electronic devices, cellulose paper-based electronics typically exhibit comparable performances in terms of the sensitivity and signal quality (i.e., signal-to-noise ratio) since similar active materials are generally used in both paper-based and traditional wearable electronics. This can be exemplified by paper-based electrophysiological sensors, which exhibit negligible difference in signal quality with conventional gel electrodes (Xu et al., 2020b). In addition, surface modifications with hydrophobic moieties can substantially improve the stability of paper-based wearable devices. However, their long-term stability and robustness under wetting environments and repeated mechanical deforma- tions are rarely studied, which are worth further investigations.

CHALLENGES AND CONCLUSION

Given the promising properties of cellulose paper in terms of inexpensive and scalable manufacturing, biodegradability, disposability, biocompatibility, breathability, and rich surface chemistry, a variety of pa- per-based devices have been developed, ranging from sensing, energy storage and generation, optoelec- tronics, and soft actuators to wireless data communication. These research advancements are driven by the attempt to seek for a more sustainable, disposable, and environmentally friendly alternative of currently polymer-based flexible electronics. Although most current studies use pristine cellulose paper as the sup- porting substrate, its abundant surface hydroxyl groups and intra- and inter-molecular hydrogen bonds offer many exciting opportunities for hybridization with other functional moieties to meet requirements of customized applications. These modifications generally contribute to the enhanced mechanical strength, decreased surface roughness, and improved thermal and chemical stability against inorganic and organic solvents.

Although a broad range of proof-of-concept paper-based wearable devices have been demonstrated, many challenges still remain, especially for the transition from laboratory-based demonstrations to prac- tical uses. These obstacles involve material and chemistry aspects, which are critical for cellulose paper as a sustainable alternative of polymer substrates. For example, although the hierarchically porous struc- ture formed by the entangled fibrous cellulose network offers excellent breathability for skin-interfaced electronics and large surface area for energy storage-related applications, the porosity inevitably results in the incompatibility with some solution-printing techniques (e.g., inkjet printing) owing to uncontrolled ink spreading. This will further cause conductivity deterioration as a result of ink penetration into a deeper

depth of the paper. Moreover, as compared with conventional miniaturized electronic devices, the printing resolution on cellulose paper has yet met the demand for future highly integrated electronics. In addition, the microscale surface roughness and intrinsic opaque nature of commercial cellulose paper limit its appli- cation in optoelectronics that typically requires high optical transmittance and low surface roughness. Un- like conventional polymers, pristine cellulose paper is not mechanically or thermally stable. Dimensional deformations caused by moisture absorption will compromise device performance and the reliability of the recorded data.

Although there are multiple options available in the market, adhesiveness is of superior importance for skin-interfaced wearable electronics since conformal contact with human skin is a critical factor of high-fi- delity biosignal recording. Motion-induced artifacts during regular human activities will cause increased noise level and signal degradation. Examples of adhesive papers used for wearable electronics include Post-it Note paper and water-soluble adhesive paper. The stickiness is often associated with a thin layer of adhesive polymer, which comes at the sacrifice of breathability to some extent. This issue can probably be addressed by innovative material design that can provide the required adhesiveness while simulta- neously preserving the porous structure. While adhesive paper is commercially available, the deposition of conductive electrodes that are typically non-sticky will compromise the overall interface adhesion. Adopting an open-mesh serpentine layout is an effective strategy since this can maximize the proportion of nonconductive adhesive area and minimize deformation-induced local strains. Alternatively, it is desir- able to develop adhesive conductive composites, comprising conductive fillers and adhesive polymers, as sensing electrodes.

In the light of an eco-friendly society, paper-based wearable electronics will be completely disposable when biodegradable electronic materials are used for device fabrication. Although various emerging carbonaceous nanomaterials bring promising routes toward paper-based all-biodegradable device, the performance is yet comparable with conventional nonbiodegradable materials. Using some nonbiode-gradable components is still indispensable for high-performance tasks, such as data processing and man- agement. Besides, the research of paper-based, highly integrated, wearable electronic system is still in the infant stage, which requires more exploration of fundamental science and technologies at the molecular level and all-paper based system assembly.

These obstacles inevitably impede the advancement and commercialization of cellulose paper-based wearable electronics. Nevertheless, they can be addressed through judicious material innovation, struc- tural engineering, and advancement of other related realms (e.g., fabrication techniques) to satisfy specific demands for various arising applications. For example, stability issues in wet environments can be allevi- ated through proper chemical functionalization (e.g., silanization); substantial surface roughness and low transmittance can be resolved by downscaling constituent fibrous diameters; inadequate adhesion to skin can be enhanced by hybridizing cellulose paper with adhesive chemicals or proper bioinspired struc- ture engineering (e.g., bioinspired dry adhesive structure [Chun et al., 2018; Wang, 2018]); low stretchability can be improved by introducing kirigami design. In addition to conventional copy paper, an expanded array of nanopapers comprising nanofibrillated cellulose is emerging thanks to extensive research efforts on fundamental research studies in terms of materials and chemistry innovations. These nanopapers have raised substantial interests as they bypass existing issues of microscale cellulose counterparts, such as considerably improved surface roughness and transparency. However, microscale and nanoscale cellulose papers are only negligibly stretchable, which might limit their wide adoption in wearable electronics. Nevertheless, we anticipate fertile opportunities of cellulose papers in sustainable wearable electronics considering their mechanical, economic, and environmental benefits.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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