iScience

CellPress OPEN ACCESS

Review

Flexible organic optoelectronic devices on paper

Teng Pan,[1] Shihao Liu,[1] Letian Zhang,[1,*] and Wenfa Xie[1,*]

SUMMARY

Paper substrate has many advantages, such as low cost, bendable, foldable, printable, and environmentally friendly recycling. Nowadays, paper has been further extended as a flexible platform to deliver electronic information with the integration of organic optoelectronic devices, such as organic thin-film transistor, organic solar cell, organic electrochromic device, and organic light-emitting device. It has great potential to become the new generation of flexible substrate. Given rough surface and porous of paper, many efforts have been underway in recent years to enable the compatibility between optoelectronics and paper substrate. In this review, we present the development history of paper and its physicochemical properties, and summarize the current development of paper-based organic optoelectronic devices. We also discuss the challenges that need to be addressed before practical uses of paper-based organic optoelectronic devices.

INTRODUCTION

As one of the greatest inventions in human history, paper has existed for more than two thousand years (Brusso, 2021; Peng et al., 2019). Paper has played an irreplaceable role in human society as the most important information carrier for mankind. Nowadays, paper often appears in scientific research (Sarrazin et al., 2007; Zakaria et al., 2005). Many of these studies want to use a "printing-like" process to print various materials on paper to prepare electronic devices (Brody, 1984; Tobjork and Osterbacka, 2011).

With the increasing demand for product diversity, paper substrates are increasingly applied in organic optoelectronic devices, such as organic thin-film transistor (OTFT), organic solar cell (OSC), organic electrochromic device (OECD), and organic light-emitting device (OLED). Compared with traditional inorganic materials, organic materials have good flexibility and are more suitable for flexible substrates such as paper substrates (Lee et al., 2019; Lim et al., 2021; Song et al., 2021; Sun et al., 2019; Xu et al., 2021). As a substrate for flexible electronic devices, paper has many unique advantages. Cellulose, the raw material fiber of paper, is the most abundant renewable and biodegradable polymer on earth (Hoeng et al., 2016; Reimer and Zollfrank, 2021). Paper is inexpensive. The price of paper (0.1 cent dm^{-2}) is substantially lower than plastic (Tobjork and Osterbacka, 2011). Furthermore, the application of paper substrates in flexible optoelectronic devices can undoubtedly alleviate the negative effects of electronic waste.

Up to now, there has been a lot of research of paper-based organic optoelectronic devices. But unfortunately, the performance of paper-based organic optoelectronic devices is limited by the inherent characteristics of the paper substrate (Hyun et al., 2013; Lee et al., 2014; Weng et al., 2020). The rough surface of the paper substrate is likely to cause a large leakage current of the device, resulting in a substantial decrease in the performance of the device. In addition, paper has poor insulation against chemicals, moisture, and oxygen, reducing the performance and reliability of the device. Nowadays, many research studies have turned their attention to the synthesis and application of new paper substrates, such as cellulose nanocrystal (Droguet et al., 2021; Rahman and Rimu, 2020) (CNC) substrate, cellulose nanofibrils (Amini et al., 2020; Solala et al., 2020) (CNF) substrate, and regenerated cellulose (Tu et al., 2021; Wang et al., 2021) (RCF) substrate. These paper substrates all retain the advantages of the commercial paper substrates. Besides, novel paper exhibits excellent characteristics, such as high transparency and smooth surface morphology (Isogai, 2021). The emergence of novel paper makes the application of paper substrates in flexible organic optoelectronic devices more reliable.

In this review, we focus our attention on these reports and sort out the relevant research on paper-based organic optoelectronic devices in the past few years. The development of paper since it was invented is discussed in development of paper section. In the paper-based flexible organic optoelectronic devices

[1] State Key Laboratory of Integrated Optoelectronics, College of Electronics Science and Engineering, Jilin University, Changchun 130012, China

*Correspondence: zlt@jlu.edu.cn (L.Z.), xiewf@jlu.edu.cn (W.X.)

https://doi.org/10.1016/j.isci.2022.103782

section, we introduced the development of paper-based organic optoelectronic devices and their applications, including OTFT, OSC, OECD, and OLED. Finally, a conclusion and outlook are given in challenges and conclusion section.

DEVELOPMENT OF PAPER

Development history

As the long-term record carrier of human civilization, paper has played a vital role in human history. We believe that the paper was first made by China's Cai Lun in AD 105. The materials used were bark, hemp, rags, and fishing nets. Later, with the gradual improvement of papermaking technology, the output of paper gradually increased, and it was used more in human daily life (Alava and Niskanen, 2006; Hubbe and Bowden, 2009; Peng et al., 2019; Robinson, 2013). As a result, a large number of books were introduced to the public, which enabled more people to receive education and greatly promoted the development of human society. After that, the network developed rapidly. As a record carrier, electrical appliances and the Internet have more powerful capabilities than paper, but paper still plays an important role in recording. In recent years, paper has many applications in research field. At the same time, many new papermaking methods have emerged, combining cellulose with some other materials, directional modification of paper under the premise of giving play to the inherent advantages of paper, and applying it to various researches. Paper has lasted for two thousand years in human history. It's supposed that within the foreseeable hundreds of years, paper will still be one of the important medium for the record of human civilization, and will play an increasing role in other scientific research fields (Han et al., 2020; Hasan and Walia, 2021; Jain and Gupta, 2021; Zhong et al., 2019).

Chemical compositions of paper

Cellulose is the original raw material of paper, and its schematic diagram is shown in Figure 1(Reimer and Zollfrank, 2021; Tobjork and Osterbacka, 2011; Zhu et al., 2013a).Conventional paper is made by dewatering a dilute suspension of cellulose fibers, and the filtration process is followed by pressing and heating.

In recent years, a variety of nanopaper is also used in experiments. Ummartyotin et al. used fibrillated bacterial cellulose and polyurethane (PU)-based resins to synthesize the nanocomposite film cellulose substrates (Ummartyotin et al., 2012). The visible light transmittance of the nanocomposite film is as high as 80%. Its thermal stability is up to 150°C. Zhu et al. focused on cellulose-based transparent, biodegradable substrates incorporating either nanopaper or an RCF (Zhu et al., 2013b). And the picture of RCF is shown in Figure 2A. Similarly, Zheng's group prepared an RCF by stirring at a constant temperature (110 ± 1°C) for 4 h to obtain a uniform cellulose-NMMO solution (Zheng et al., 2021). The cellulose-NMMO solution was coated on a clean glass substrate to form a cellulose film. As shown in Figure 2B, Chen et al. prepared a CNC/colorless polyimides (CPI) hybrid substrate with high optical, mechanical, and thermal properties (Chen et al., 2020). CNC has an excellence performance, with the thermal decomposition temperature of 555°C and folding capacity of 160,000 times. Particularly, the substrate keeps an excellent transmittance of 86% at 600 nm, and it is colorless. The schematic illustration of the preparation of cellulose nanofibrils and polyarylate (CNF/PAR) hybrid substrate are shown in Figure 2C, which was achieved by Tao's group (Tao et al., 2020). In addition, cellulose and its derivatives can also be used for a wider range of purposes, such as interlayers and additives of electronic device(Liu et al., 2020b; Wu et al., 2020b).

Physicochemical properties of paper

Surface roughness

The material placed on substrates with high roughness is likely to form similar surface morphology, which leads to uneven device thickness. Moreover, the leakage current of the device on rough substrates will increase and thus reduce the working efficiency. Generally speaking, because commercial paper has loads of spikes and burrs on its surface, its surface roughness is quite high. In contrast, the new nanopaper substrate adopts different synthesis methods, and has denser cellulose arrangement, thus resulting in relatively smoother surface.

Voggu et al. characterized six kind of paper by SEM, including office paper and bacterial cellulose (Voggu et al., 2017). He's group characterized surface morphology of tracing paper and printing paper (He et al., 2020). The average root-mean-square surface roughness determined by AFM analysis for tracing paper is 243 nm, which is smoother than that of printing paper, with a surface roughness of 482 nm (Figure 3A).

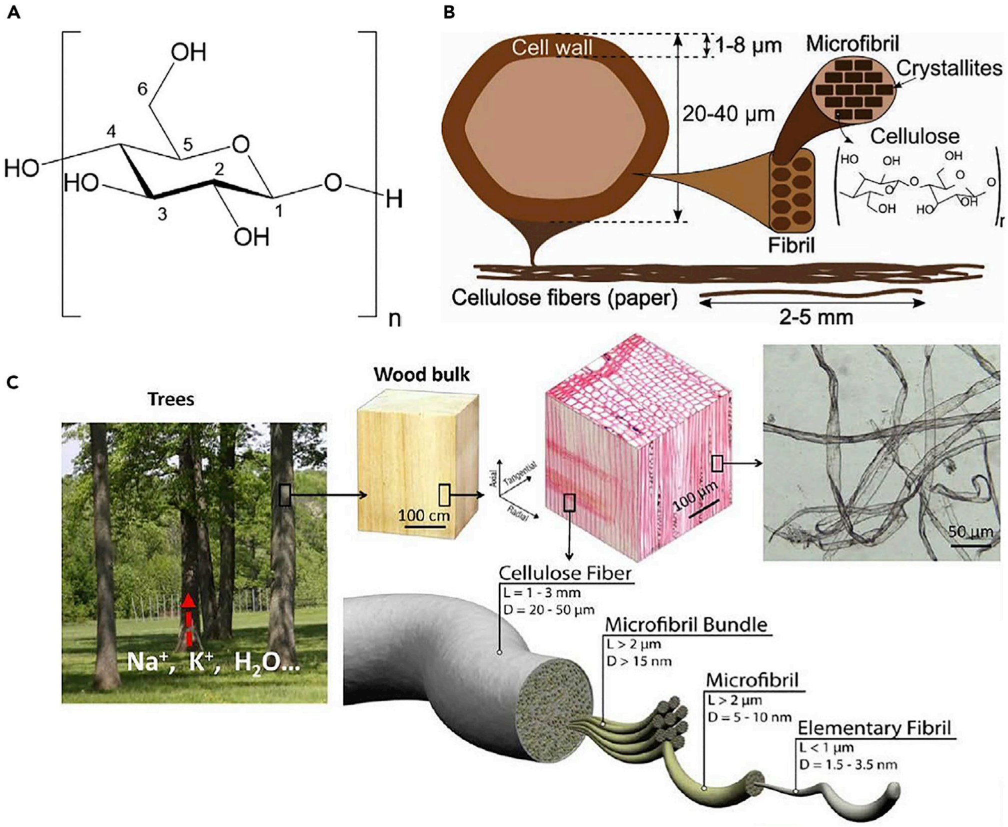

Figure 1. Schematic diagram of cellulose

(A) Molecular structure of cellulose. Adapted with permission from (Reimer and Zollfrank, 2021).Copyright 2021, Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim.

(B) The structure of the cellulose fibers. Adapted with permission from (Tobjork and Osterbacka, 2011).Copyright 2011, Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim.

(C) Hierarchical structure of wood fiber. Adapted with permission from (Zhu et al., 2013a).Copyright 2013, American Chemical Society.

Jin' group characterized chitin nanofibers (ChNF) by AFM and SEM. The building blocks of ChNF paper are ultrafine nanofibers composed of chitin molecules (Jin et al., 2016). Jiang et al. characterized nanopaper composited by different methods (Jiang et al., 2020). The small size of the lignocellulose nanofibrils coupled with the cementing effect of lignin results in the highly dense structure of lignocellulose nanopapers.

Mechanical properties

The flexible substrate serves as the support of the organic optoelectronic device, which plays a decisive role in the flexible surface of the device. Compared with plastic substrates, paper can be folded, have a smaller radius of curvature, and can be twisted. The mechanical properties of paper substrates are usually characterized by parameters such as stress-strain curve, elastic modulus or Young's modulus. Jin's group displayed the engineering stress-strain curves of the ChNF and copy paper by tensile testing (Jin et al., 2016). The elastic modulus of ChNF (4.3 GPa) is comparable to that of the PI film (3.9 GPa). The effects of fiber orientation and laminate stacking sequence on the mechanical anisotropy of paper were examined by Kroeling (Kroeling et al., 2018). The stress-strain curves for the substrates of paper, plastic paper, and epoxy resin film were measured by Hu's group, the results are shown in Figures 3B and 3C (Yao et al., 2016). And the ultimate tensile strength and Young's modulus of the paper-based all-cellulose composite laminates were 191 MPa and 17.5 GPa in the fiber direction, respectively, compared with 104 MPa and 10.4 GPa in the transverse direction, respectively. Hall et al. investigate various bend tests and apply these tests to aged paper with the goal of finding easy, non-destructive tests to determine the mechanical properties of paper (Hall et al., 2019).

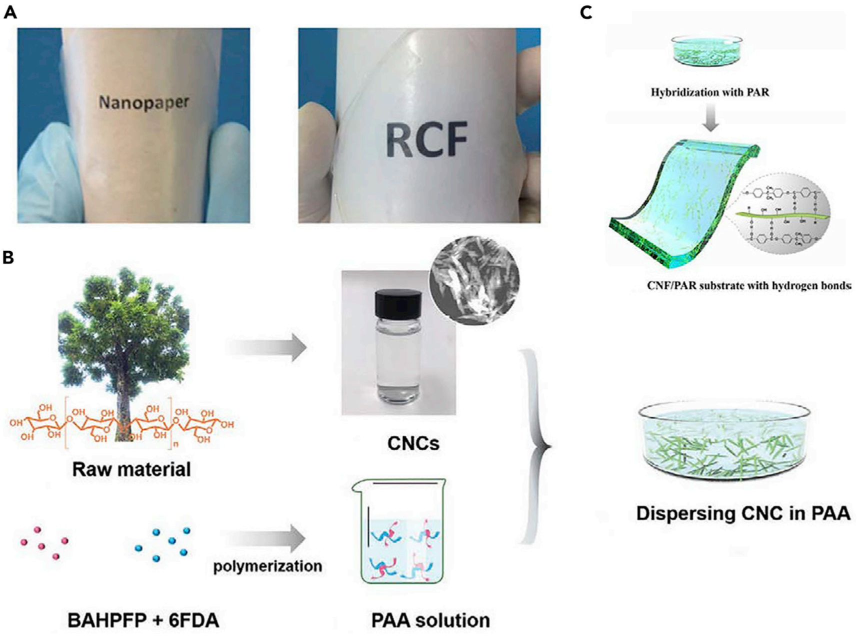

Figure 2. Schematic diagram of the synthesis process of the novel nanopaper

(A) Digital images showing the transparency and flexibility of the nanopaper and RCF. Adapted with permission from (Zhu et al., 2013b).Copyright 2013, Royal Society of Chemistry.

(B) Schematic illustration of the preparation of cellulose nanocrystal and colorless polyimide (CNC/CPI) hybrid substrate and its application in OLED fabrication. Adapted with permission from (Chen et al., 2020).Copyright 2020, Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim.

(C) Schematic illustration of the preparation of cellulose nanofibrils and polyarylate (CNF/PAR) hybrid substrate. Adapted with permission from (Tao et al., 2020).Copyright 2020, American Chemical Society.

Optical properties

For organic optoelectronic devices, the transmittance of the substrate is also very important, especially for optoelectronic devices and issues related to the direction of light incidence and exit. The optical haze is also one of the important optical properties (Yao et al., 2016). The transmittance and transmittance haze of plastic (PET), roll paper, and plastic paper are measured by Hu's group (Figures 3D and 3E). To increase the efficiencies of light coupling into and out of optoelectronic devices, the high transmittance and high haze is desired. Zhu's group displayed transmittance and optical haze of different substrate (Zhu et al., 2013b). Indicating that the nanopaper has the highest diffusive transmittance but the lowest specular transmittance, the value is up to 50% for nanopaper and for RCF it is much less in most of the wavelength range. Purandare's group measured the optical transmission of cellulose film (Purandare et al., 2014). As a result, the thicker 40-μm cellulose film had a transmission of 83% at 400 nm increasing to 86% at 850 nm. Pinto et al. prepared a transparent composites substrate by using bacterial cellulose (BC) and castor oil based-polyurethane (PU) (Pinto et al., 2015). And the composites substrate showed a great transmittance, the value higher than 90% in the visible region.

Thermal properties

During the operation process, the extra heat vastly affects the service life of the organic optoelectronic device. High-temperature heating causes severe damage to substrate. Therefore, thermal stability is very important for flexible substrate. Compared with plastic substrates such as PET, both commercial paper and nanopaper have a lower coefficient of thermal expansion and a higher thermal decomposition temperature. Hsieh et al. fabricated flexible electronics on highly heat-resistant nanopaper substrate (Hsieh et al., 2014). The thermal gravity (TG) curves for the hardwood cellulose pulp paper and the cellulose nanopaper were compared. Both paper substrates were heated up to 600°C, neither substrate showed a glass transition point, and thermal degradation did not occur in either substrate until approximately 300°C. Nogi's group reported a cellulose nanofiber paper with an extremely low CTE of 5–8 ppm/K (Nogi et al., 2013). During 150°C heating, the total transmittance was constant at 87.4%–88.1% (Figure 3F), while the haze increased linearly from 4.5% to 14.0%.

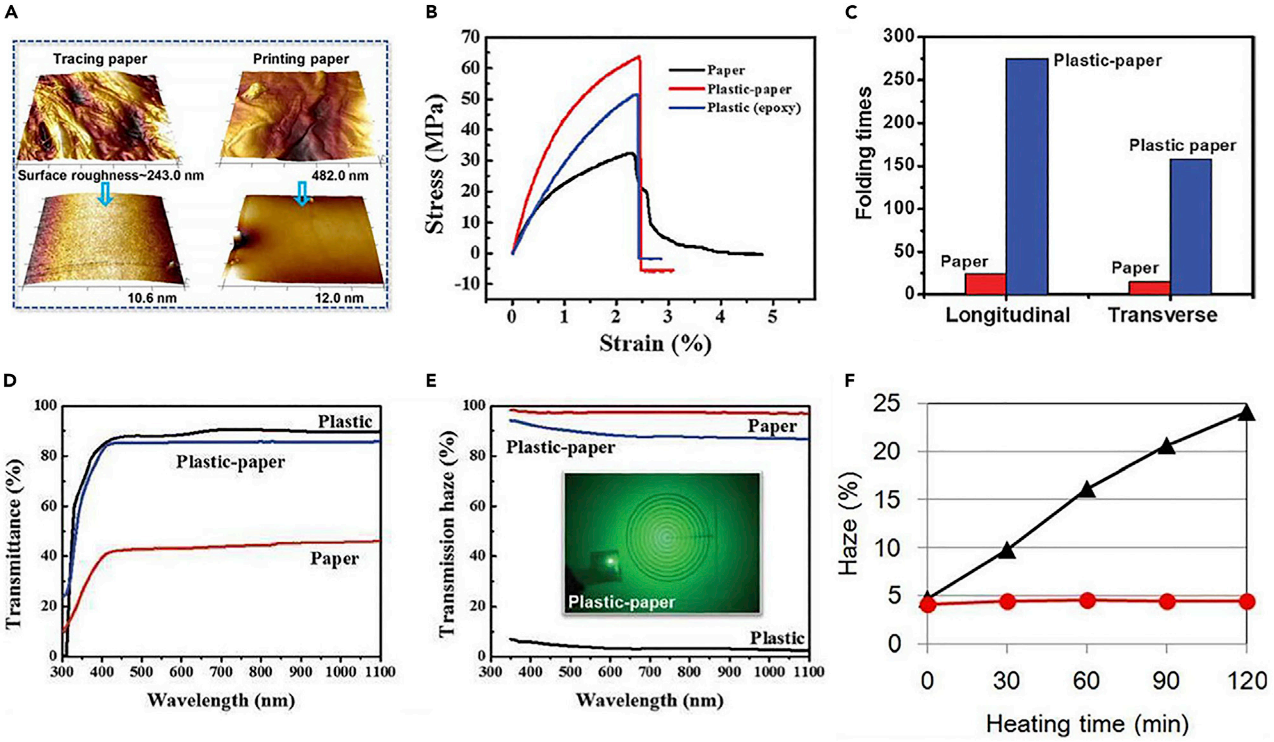

Figure 3. Physicochemical properties of paper

(A) AFM images of tracing paper and printing paper. Adapted with permission from (He et al., 2020). Copyright 2020, American Chemical Society.

(B) Mechanical stress-stain curves for paper, plastic paper, and epoxy resin film. Adapted with permission from (Yao et al., 2016). Copyright 2016, Royal Society of Chemistry.

(C) Folding capacity measurement of roll paper and plastic paper in the longitudinal direction and transverse direction. Adapted with permission from (Yao et al., 2016). Copyright 2016, Royal Society of Chemistry.

(D) Total transmittance of plastic (PET), roll paper, and plastic paper. Adapted with permission from (Yao et al., 2016). Copyright 2016, Royal Society of Chemistry.

(E) Total transmittance haze of plastic (PET), roll paper, and plastic paper. Adapted with permission from (Yao et al., 2016). Copyright 2016, Royal Society of Chemistry.

(F) Total light transmittance during heating at 150°C for 120 min. Adapted with permission from (Nogi et al., 2013).Copyright 2013, American Institute of Physics Publishing.

Moisture and oxygen transmission rate

For organic optoelectronic devices, the moisture and oxygen barrier properties of the substrate are very critical. Compared to inorganic materials, organic materials are extremely sensitive to moisture and oxygen. Therefore, the moisture and oxygen transmission rate of the substrate is very important to the working life of the device (Ji et al., 2020; Song et al., 2014). Common flexible substrates for organic optoelectronic devices, such as PET and conventional paper substrates, usually have a low moisture and oxygen transmission rate, which is far from glass substrates. Even the nanopaper substrate synthesized through the optimized process, its moisture and oxygen transmission rate is difficult to reach the same level as that of the glass substrate. Therefore, the surface of the device needs to be modified with some moisture-blocking materials before the device is prepared. Or the device is packaged after the device is prepared to increase the service life of the device (Kang et al., 2020).

Modification processes of paper substrate

For commercial paper substrates, it must undergo some processing before it can be applied to flexible optoelectronic devices. The spin coating method is one of the most common solution methods. Barsotti's group process tattoo paper by spin coating (Figure 4A) (Barsotti et al., 2021). They deposited PMMA on tattoo paper as a buffer layer. The thickness of the PMMA buffer layer was about 1.5 μm and the roughness was less than 2 nm. Wang et al. fabricated OFETs based on the art paper substrates (Xu et al., 2019). On the

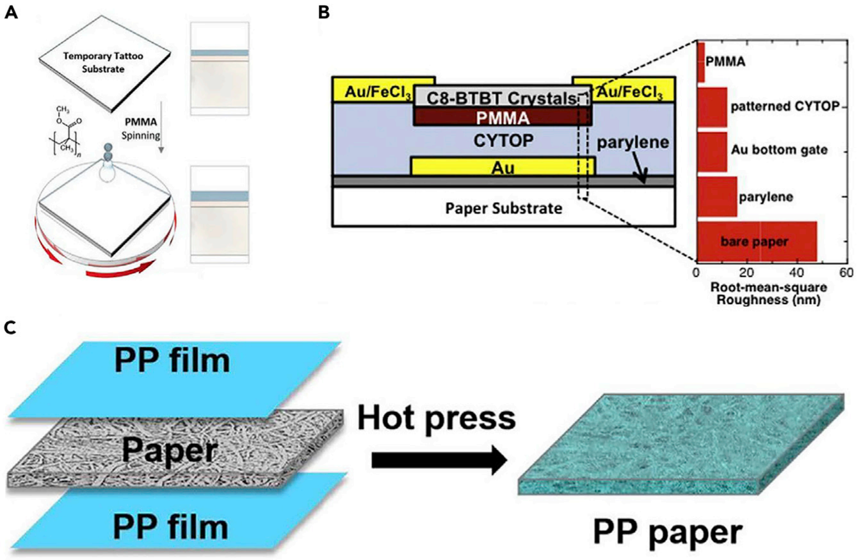

Figure 4. Schematic diagram of modification processes of paper substrate

(A) Spin coating process of tattoo paper. Adapted with permission from (Barsotti et al., 2021). Copyright 2021, Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim.

(B) Schematic diagram of flexible OTFTs fabricated on the paper substrate. Adapted with permission from (Li et al., 2012). Copyright 2012, Elsevier.

(C) Schematic of the fabrication of composite PP paper via hot press. Adapted with permission from (Huang et al., 2019).Copyright 2019, Elsevier.

surface of the paper substrate, CL-PVP layer was deposited by spin coating as a buffer layer. A thin film with a smooth surface can be prepared by spin coating, but in most cases the films are too thin for paper. Chemical vapor deposition is a technology which mainly uses gas phase compounds to produce thin films. Yoon's group used chemical vapor deposition to prepare 6-μm thick parylene and 100-nanometer thick SiO_2 on a common printing paper substrate (Yoon and Moon, 2012). The 6-μm thick parylene serves as the flat layer of the substrate, while the role of SiO_2 is to enhance the moisture and oxygen barrier of the substrate. Li et al. prepared 3 μm of parylene on a paper substrate as a moisture and oxygen barrier, and the surface roughness of the substrate was also reduced after depositing parylene (Figure 4B) (Li et al., 2012). Although dense films can be produced, the disadvantage of CVD is its higher cost. Hot pressing is a simple mechanical method that presses multiple layers of materials together. Yang et al. prepared the novel paper substrate through a simple hot-press process with commercial paper and polypropylene (PP) film (Yang et al., 2020). Then, kinds of organic optoelectronics were fabricated based on the novel substrate. Hwang et al. fabricated a dry film photoresist (DFR) on paper by hot press (Kim and Hwang, 2018). The DFR acts as a planarization layer and negative photoresist to realize the pattern of OLED. Wang et al. combine the polypropylene (PP) film and printing paper and fabricated paper-based OLEDs and paper-based OSCs (Figure 4C) (Huang et al., 2019).

PAPER-BASED FLEXIBLE ORGANIC OPTOELECTRONIC DEVICES

Organic thin-film transistors

Transistor is a solid semiconductor device with multiple functions such as detection, rectification, amplification, switching, voltage stabilization, and signal modulation. As a variable current switch, the transistor can control the output current based on the input voltage. Organic thin-film transistors (OTFTs) have been demonstrated an excellent performance (Deng et al., 2021; Kim et al., 2020, 2021; Lu et al., 2020; Makita et al., 2021).

In term of the device structure, OTFTs can be divided into a bottom-gate structure, top-gate structure, and in-plane gate structure. The bottom-gate structure is the most traditional structure. Kraft et al. demonstrated bottom-gate OTFTs on the surface of banknote (Kraft et al., 2019). They fabricated OTFTs in same structure on glass, PEN, and banknote (Figure 5A). As the surface roughness of the device substrate

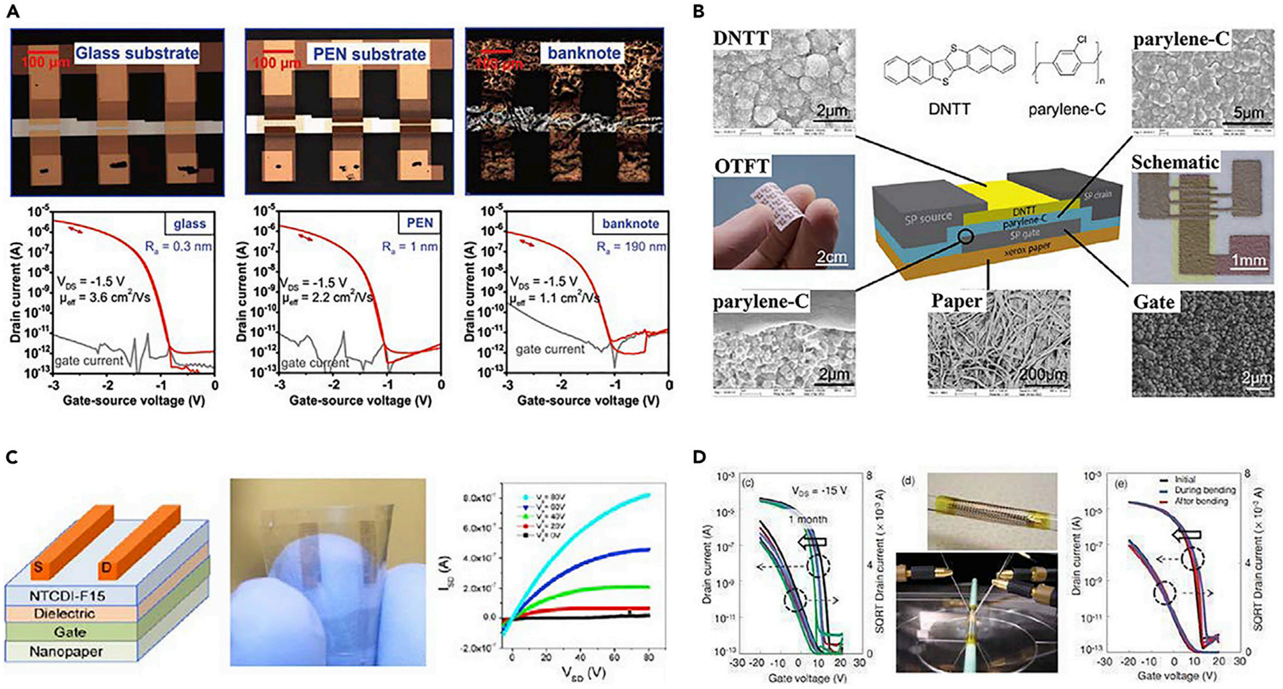

Figure 5. The performance and schematic diagram of bottom-gate OTFT

(A) Photographs and transfer characteristics of p-channel DNTT TFTs fabricated on glass, PEN, and a banknote. Adapted with permission from (Kraft et al., 2019). Copyright 2018, Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim.

(B) Transistor structure and electrical performance. Adapted with permission from (Peng and Chan, 2014). Copyright 2013, Elsevier.

(C) The picture and performance of paper-based OFETs. Adapted with permission from (Huang et al., 2013). Copyright 2013, American Chemical Society.

(D) Typical electrical performances of fabricated OTFT array. Adapted with permission from (Fujisaki et al., 2014).Copyright 2013, Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim.

increases, the effective mobility gradually goes down. It can be attributed to the detrimental effect of the surface roughness on the degree of molecular ordering in the organic semiconductor film. Paper-based OTFTs with great memory properties were fabricated by Chan's group (Figure 5B) (Peng and Chan, 2014). All contact electrodes were printed on standard untreated Fuji Xerox printer paper without using planarization layer. The screen-printed gate electrodes play an important role in suppressing the surface roughness of the substrate, and the averaged leakage current of the device is around 10 pA. Otherwise, the transistors show average mobility of 0.297 $cm^2 V^{-1} s^{-1}$ and on/off ratio larger than 10^5. The paper-based OTFTs also show a reliable memory retention time for more than 10,000 s, because the extraordinary stable traps state in the paper dielectric. High-performance operationally stable OTFTs on a PowerCoat HD 230 paper were demonstrated (Raghuwanshi et al., 2019). The PVA was deposited on the surface of paper substrate by spin coating as a buffer layer and barrier layer. The paper-based OTFTs show excellent electrical properties, with a maximum and av field-effect mobility of 0.44 and 0.22 (±0.11) $cm^2 V^{-1} s^{-1}$, respectively. Furthermore, these devices displayed very stable electrical characteristics after long exposure periods to humidity and an excellent shelf life of more than 6 months in ambient environment.

Nanopaper is often used in paper-based OTFTs as well. Hu et al. fabricated OFETs on the transparent nanopaper substrate (Huang et al., 2013). The picture of paper-based OFETs is shown in Figure 5C. Fujisaki et al. demonstrated the OTFT array on transparent nanopaper substrate (Figure 5D) (Fujisaki et al., 2014). A short-channel bottom-contact OTFT is successfully fabricated on the nanopaper by a lithographic and solution-based process. The paper-based OTFTs exhibit a high hole mobility of up to 1$cm^2 V^{-1} s^{-1}$ and a small hysteresis of below 0.1 V under ambient conditions. Pentacene-based OTFTs were fabricated on commercial photo paper, ultra-smooth specialty paper, and ultra-thin (100 µM) flexible glass by Steckl's group (Zocco et al., 2014). The paper was used as substrate without planarization layer. All function layers were deposited by dry step process. Parylene C acts as the insulator layer and pentacene acts as semiconductor materials. The results on the lifetime of OTFTs on photo paper yielded stable transconductance and mobility values over a period of more than 250 h.

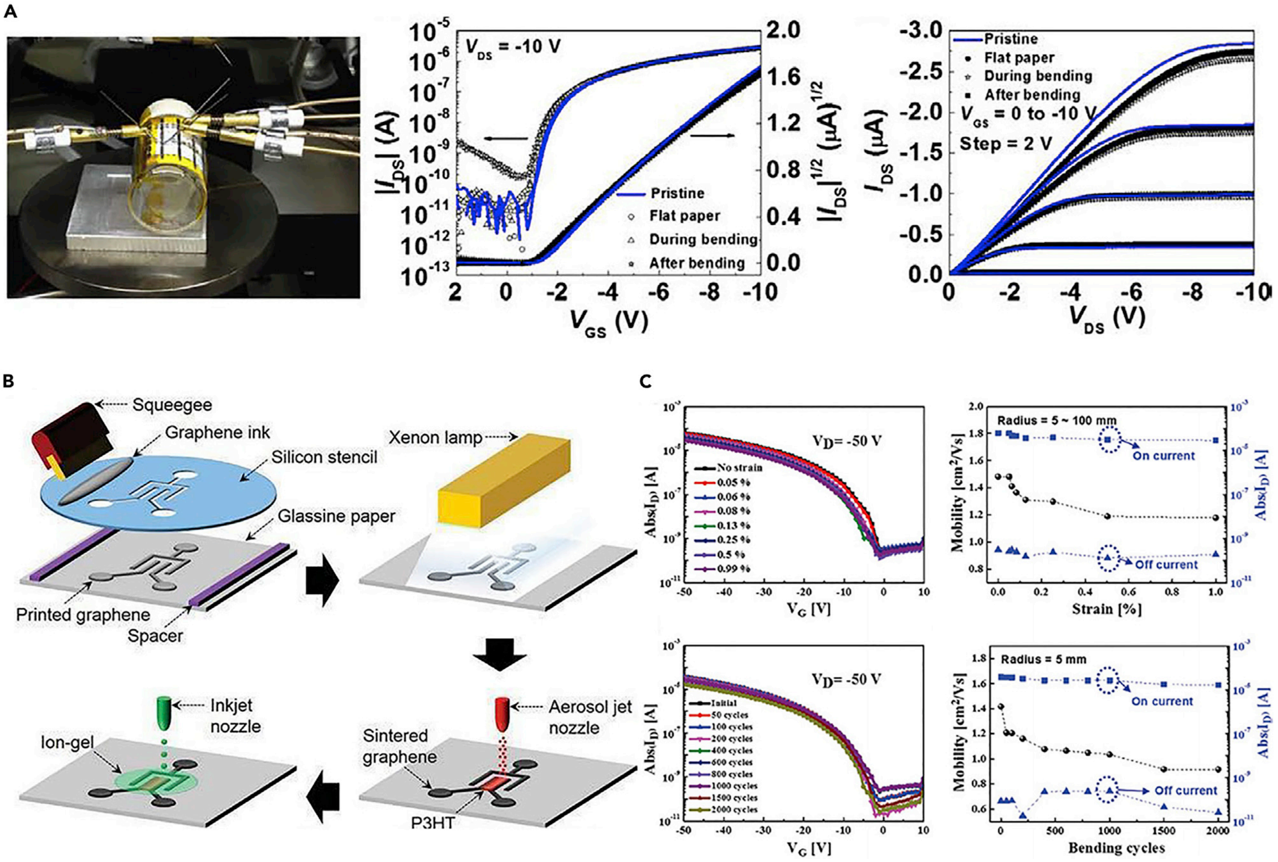

Figure 6. The performance and schematic diagram of top-gate OTFT, in-plane gate OTFTs, and OPTs

(A) Photograph and characteristics of OFET in the bending test. Adapted with permission from (Wang et al., 2017). Copyright 2016, Elsevier.

(B) Fabrication steps for an all-printed organic TFT on a glassine paper substrate. Adapted with permission from (Hyun et al., 2015). Copyright 2015, Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim.

(C) Transfer characteristics and μ_{sat} at different state. Adapted with permission from (Park et al., 2018).Copyright 2018, Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim.

To date, most of the research on OTFTs uses a bottom-gate device structure. The organic film of the top-gate structure is prepared on the source and drain electrodes. In term of technological, the preparation process of the source and drain electrodes may have an adverse effect on the performance of the organic film that has been formed, and the top-gate structure does not have this problem. Kippelen et al. reported on top-gate OFETs based on specialty paper (Wang et al., 2017). The mixture of PVA and PVP was spin coating on paper as planarization layer. The gate dielectric can be engineered to enable top-gate OFETs to operate at low voltage values and to display exceptional operational and environmental stability even in aqueous environments. Top-gate OFETs fabricated on HD 230 paper display low threshold voltage values in the range of −2 to −3 V and high carrier mobility values in the range of 0.1–0.4 cm^2 V^{-1} s^{-1}. OFETs based on cellulose nanocrystal−glycerol (CNC/glycerol) substrates were also demonstrated by Kippelen's group (Figure 6A) (Wang et al., 2015). Al_2O_3 layer was produced on CNC substrate by atomic layer deposition (ALD), acts as a passivation layer and planarization layer. They fabricated the OTFTs without the passivation layer (D1), and a normal device (D2), without the passivation layer, the mobility values decrease from 0.23 cm^2 V^{-1} s^{-1} to 0.13 cm^2 V^{-1} s^{-1}. Furthermore, in the D1 OFETs, the mobility decreased to a value of 0.08 cm^2 V^{-1} s^{-1}, whereas in the D2 devices, the mobility remained unchanged with a value of 0.23 cm^2 V^{-1} s^{-1}. The decreased mobility in the D1 devices suggests that chemical or physical interactions occur between the substrate and the organic semiconductor. And the result of X-ray photoelectron spectroscopy (XPS) measurements identified the chemical or physical interactions between the substrate and the organic semiconductor.

In addition to the bottom-gate structure and the top-gate structure, in-plane gate structures are also used in OTFTs. Sun et al. fabricated an in-plane gate OTFTs on cellulose paper. The biocompatible chitosan is used as the smoothening layer, and laminated ion gel is used as high capacitance dielectrics (Qian et al., 2015). The laminated ion gels can be *in situ* peeled off and reused in other FETs, and the device performance is not degenerated obviously with repeated use. The current on/off ratio and V_{th} values of all the devices gated with the reused ion gels are not degenerated obviously with the times of repeated use, which indicates that the ion gels peeled off by tweezers can be reused in OFETs. Frisbie's group fabricated all-print OTFTs on glassine paper (Hyun et al., 2015). Graphene electrodes were deposited by screen printing, and the gate, source, drain were fabricated in one process. The other functional layer were deposited by aerosol-jet printing. Schematic diagram as shown in Figure 6B.

OFETs are the most common type of OTFTs. In addition, there are a variety of OTFTs that can perform other functions. Ju et al. fabricated a transparent organic phototransistors (OPTs) that can detect visible light with nontoxic organic active materials on biodegradable substrates toward environment-friendly electronics (Figure 6C) (Park et al., 2018). The phototransistors, which can detect visible light and perform in two operation modes, exhibit a maximum responsivity of 54.8 A W^{-1} and a photosensitivity of 24.4 under white light illumination at an intensity of 0.12mWcm^{-2}. Meng's group demonstrated self-supported hysteresis-free flexible organic thermal transistor (OTTs) based on commercial graphite paper (Zhu et al., 2018). The paper-based OTTs exhibit extremely low hysteresis, wide operating temperature range (20°C–100°C), high stability, and temperature sensing performance. Such thermal transistors are promising for integration in current electronic devices and promote the diversity of the flexible transistor substrates.

Organic solar cells

Energy shortages and environmental pollution caused by fossil fuels have become an exigent problem to be solved, the replacement of clean and recyclable green energy such as solar energy can alleviate this problem. Organic solar cells (OSCs) are an established and optional method by effectively transforming light energy into electrical energy (Burlingame et al., 2020; Fukuda et al., 2020; Li et al., 2020; Liu and Chen, 2020; Wan et al., 2020).

Joh et al. prepared carbon nanosheets (CNSs) using an environmentally friendly cellulose precursor system (Figure 7A) (Son et al., 2020). Using CNS as transparent electrode (TE) material, the power conversion efficiency can reach up to ~1.84%. Cellulosic CNS is expected to have practical application value as TE material for various electronic devices. Kippelen used cellulose nanocrystals as the substrate of solar cells, and the power conversion efficiency of the cells made with CNC/Ag/PEI/P3HT:ICBA/PH1000-L as the solar cell device structure can reach up to 4% (Zhou et al., 2014). The fill factor can reach 0.64 ± 0.02. The performance level is almost the same as that of OSCs made on PES substrates. Nogueira's group successfully synthesized two nanocellulose substrates and realized their application in OSC (Costa et al., 2016). And for the first time, they discussed the different properties of cellulose substrate and their influence on OSC performance. The power conversion efficiency of the inverted OSC made of NFC, CNC, and glass substrate is 0.5%, 1.4%, and 3%, respectively. Hu's group introduced a new type of transparent paper substrate (Fang et al., 2014). The material is made of wood fiber, with ultrahigh optical transparency (about 96%) and ultrahigh haze (~60%), providing the best substrate design material for solar cell devices. The PCE of OSCs is enhanced by 10% through a simple lamination process. This low-cost, high-transparency, high-fog paper can be used as an excellent film to enhance the light-capturing performance of photovoltaic applications such as solar panels, roofs, or Windows.

Iyer et al. have successfully produced paper-based solar cells using commercially available smooth paper as the base of the cells (Figure 7B) (Rawat et al., 2019). Smoothing layers of polyvinyl formal (PVF) with a knife-edge coater give them an acceptable root-mean-square roughness of around 2.6 ± 0.2 nm. ITO-free paper substrate organic solar cell with the PCE of 3.37% with P3HT:PCBM blends as photoactive layer (PAL) and 6.44% with PTB7:PCBM blends as PAL was demonstrated. The P3HT, PTB7, and PCBM molecules were fabricated on commercial paper to produce an OSC device also by Iyer's group (Jayaraman and Iyer, 2020). Under the irradiation of AM1.5G light, the active area PCE of P3HT:PCBM and PTB7:PCBM modules are 2.38% and 4.23%, respectively, which is close to the active area PCE of modules using these molecules on glass and other substrates. Xinhua Ouyang et al. used the residues of tobacco stalks from agriculture and forestry to prepare a degradable, renewable, and sustainable CNP, and successfully applied it in OPV to achieve OSCs light capture and wide-angle capture (Figure 7C) (Wu et al., 2020a). Using CNP as the trap layer on the incident light side, the results show that the PCE value of the device is 16.17%.

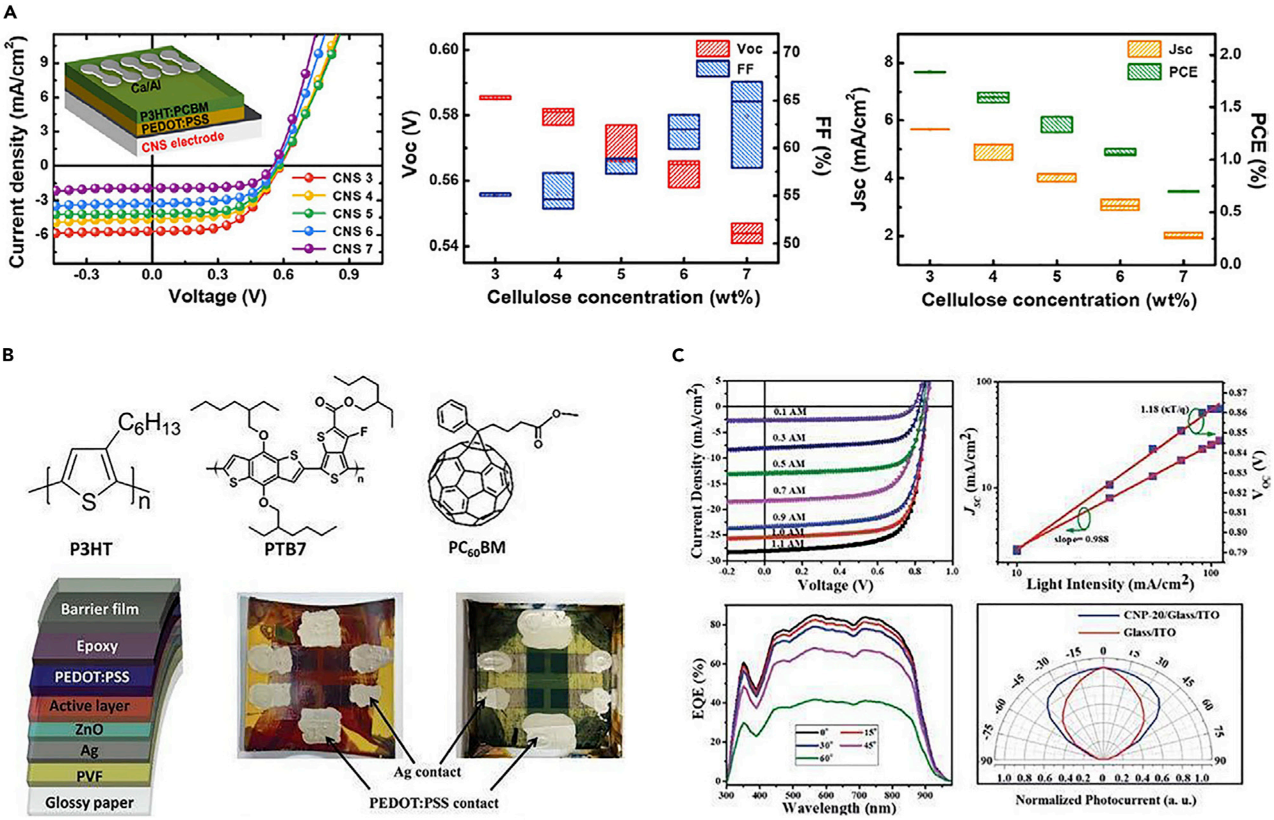

Figure 7. The performance and schematic diagram of OSCs

(A) Schematic diagram and performance of OSCs. Adapted with permission from (Son et al., 2020). Copyright 2020, Elsevier.

(B) Chemical structures of constituent molecules in PAL, schematic diagram of constituent layers in the paper solar cell device. Adapted with permission from (Rawat et al., 2019). Copyright 2019, Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim.

(C) Optical and electrical properties of OSCs. Adapted with permission from (Wu et al., 2020a).Copyright 2020, Royal Society of Chemistry.

Organic-inorganic hybrid perovskite solar cells (PSCs) are a novel and efficient optoelectronic device. Research on paper-based PSCs has also emerged in recent years. Yu et al. developed a flexible PSCs based on transparent nanocellulose paper (Gao et al., 2019). The PCE of these paper-based PSCs reached 4.25%, while the power per weight was as high as 0.56 Wg^{-1}. The flexible PSCs also showed good stability, retaining >80% of original efficiency after 50 times of bending. Song's group prepared paper-based PSCs on cellophane with a high PCE of 13.19% (Li et al., 2019). The PSCs on paper exhibit 50 single folding and 10 dual folding stability: they preserve 85.3% and 84.1% of the initial PCE after −180° and +180° single folding for 50 cycles, respectively; and they remain 67.2% and 55.3% of the initial PCE after 10 inner and outer dual folding cycles, respectively.

Organic electrochromic devices

Electrochromism is a phenomenon in which the optical properties of materials (reflectance, transmittance, absorptivity, etc.) undergo stable and reversible color changes under the action of an external electric field, and appear as reversible changes in color and transparency. Materials with electrochromic properties are called electrochromic materials, and devices made of organic electrochromic materials are called organic electrochromic devices (OECDs) (Chaudhary et al., 2020; Che et al., 2019; Dov et al., 2017; Kim et al., 2019; Sonmez et al., 2004).

Electrochromic devices are of great significance in energy-efficient buildings, Internet of Things devices, and low-cost advertising applications. Liu's group doped PEDOT:PSS with a solvent was used both as the electrode and the electrochromic functional layer for fabrication of OECDs on absorptive paper surfaces (Liu et al., 2020a). Although the color contrast decreases with the increase of doped PEDOT:PSS, the speed of color conversion is higher. The color contrast range, ΔE*, of the devices fabricated on the

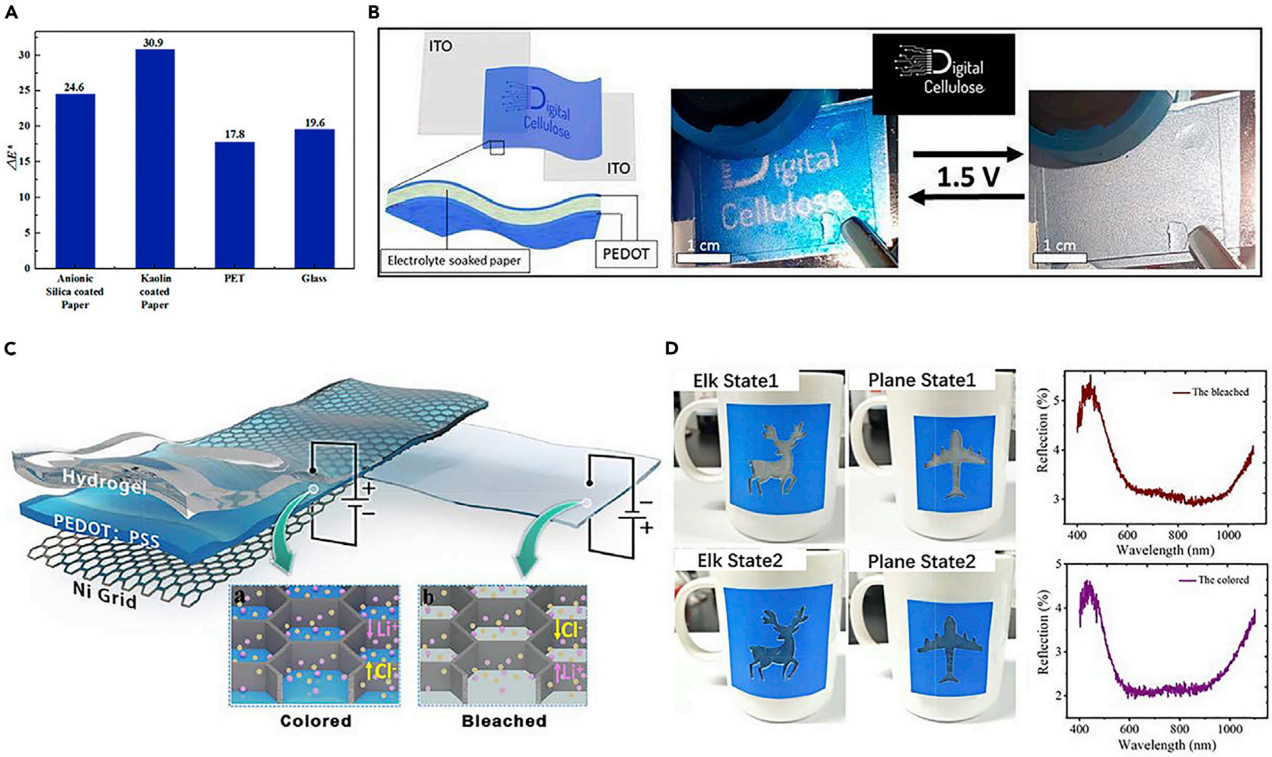

Figure 8. The performance and schematic diagram of OECDs

(A) The color contrast range, ΔE*, of the devices fabricated on the different substrates. Adapted with permission from (Liu et al., 2020a). Copyright 2020, Multidisciplinary Digital Publishing Institute.

(B) Schematics and photographs of paper electrochromic devices using electrolyte-soaked filter paper Adapted with permission from (Brooke et al., 2019). Copyright 2019, Multidisciplinary Digital Publishing Institute.

(C) Schematic illustration of the triple-layer architecture of the ultrathin ECD based on the free-standing Ni grid electrode. Adapted with permission from (Zhao et al., 2021). Copyright 2021, Optica Publishing Group.

(D) The electrochromic images of elk and plane in visible light camouflage and the reflection changes in colored and bleached ECDs among spectrum ranges of 400–800 nm. Adapted with permission from (Zhao et al., 2021).Copyright 2021, Optica Publishing Group.

different substrates is shown in Figure 8A. Jonsson et al. proposed a vapor-phase polymerized OECDs based on UV pattern and poly(3,4-ethylenedioxythiophene) film, which can switch between two high-resolution images (Brooke et al., 2019). Finally, they use a UV pattern technology to deposit electrochromic patterns on porous paper to produce functional paper, thus achieving environmentally friendly electrochromic display (Figure 8B). Designing an electrochromic device (ECD) that is extremely flexible and stretchable has been a huge challenge because conductive materials and supporting substrates physically deform and break after multiple bends. Liu et al. introduced a self-supporting metal nickel mesh electrode, which dispenses with solid or flexible polymer substrates and has excellent foldability (bending radius less than 50 μm) and stretchable performance (stretched to 117.6%), excellent electrical conductivity (sheet resistance less than 0.4 Ω sq^{-1}), high light transmittance (about 90% of the full spectrum), and ultrathin thickness (3.7 μm). A paper-thin, ultra-flexible, stretchable ECD with a total thickness of 113 μm was prepared by assembling metal electrodes, electrochromic materials, and hydrogels (Figure 8C) (Zhao et al., 2021). The flexibility and wearability of the three-layer substrate-free ECD makes it possible to use it as a future electronic product for a variety of uses, such as flexible displays, camouflage wearables, and medical monitoring. Ersman reported how such polymers and poly electrolytes can be cast together with nanofibrillated cellulose (NFC) derived from wood. The resulting films, which carry ionic or electronic functionalities, are all-organic, disposable, light-weight, flexible, self-adhesive, elastic, and self-supporting.

Organic light-emitting devices

In recent years, the application of organic light-emitting devices(OLED) in the display and lighting fields has been increasing(Liu et al., 2022; Wang et al., 2019; Yu et al., 2019; Zang et al., 2021; Zhang et al., 2019a; Xue

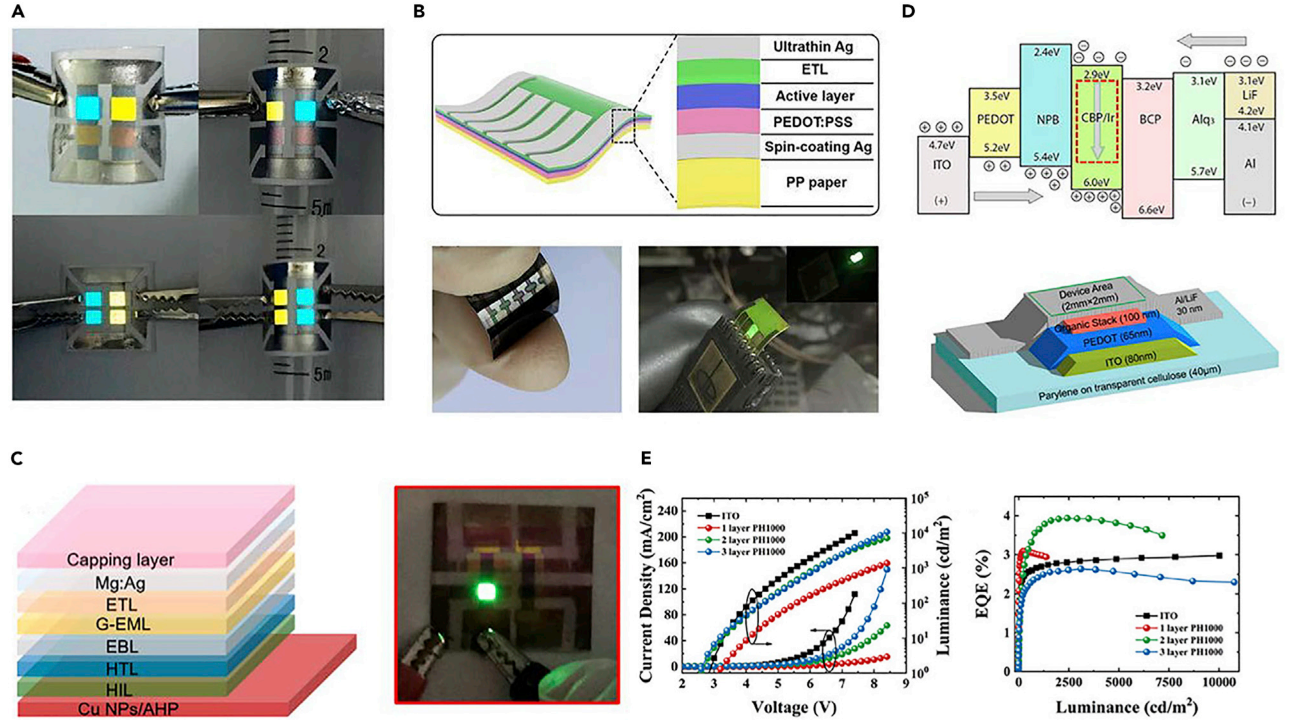

Figure 9. The performance and schematic diagram of OLEDs

(A) Pictures of DMT-OLEDs based on paper substrates. Adapted with permission from (Zhang et al., 2019b). Copyright 2019, American Chemical Society.

(B) Device architecture and picture of OLED. Adapted with permission from (Huang et al., 2019). Copyright 2019, Elsevier.

(C) Fabrication of a flexible OLED device on Cu NPs/AHP. Adapted with permission from (Yang et al., 2020). Copyright 2020, American Chemical Society.

(D) HOMO/LUMO energy levels and device structure of OLED. Adapted with permission from (Purandare et al., 2014). Copyright 2014, Institute of Physics.

(E) Performances of cellulose-based flexible green OLEDs. Adapted with permission from (Zheng et al., 2021).Copyright 2021, Royal Society of Chemistry.

and Xie, 2021; Zheng et al., 2020). Compared with conventional light-emitting devices, OLED has many advantages such as wide viewing angle, fast responding speed, and light-weight. Research of paper-based OLED generally adopted two strategies: preparing a buffer layer on a conventional paper substrate and using cellulose to synthesize a new type substrate.

The buffer layer can simultaneously reduce roughness and improve waterproof performance. A dual-micro-cavity top-emitting OLEDs (DMT-OLEDs) based on paper substrates have been fabricated by Xie's group (Figure 9A)(Zhang et al., 2019b). A cross-linked poly(4-vinylphenol) layer is deposited by spin coating and subsequently cross-linked at 160°C for 2 h, the PVP layer act as a buffer. The Ni and parylene films were sputtered on the copy paper substrates by Moon's group (Yoon et al., 2010). The parylene film is an ideal buffer layer materials because it shows very high chemical stability and excellent homogeneous and conformal coverage. The maximum luminance of the device fabricated on the paper was limited to about 1 cd m^{-2}. With the parylene film and SiO_2 coated on the both sides of the copy paper substrate, the maximum luminance of paper-based OLED reached 2200 cd m^{-2} at a driving voltage of 13 V, which was also achieved by Moon's group (Yoon and Moon, 2012). The SiO_2 is an excellent moisture resistance materials and act as a moisture-blocking layer, which is critical for OLED. Without SiO_2 layer, the maximum luminance reduced from 2200 cd m^{-2} to 81 cd m^{-2}. Ha et al. fabricated double buffer layer on paper substrate in another work (Ha et al., 2016). A 21-μm thick buffer layer was coated on both sides of copy paper substrate and the additional buffer 450-nm thick PVP layer was spin coated on one side of substrate. The maximum luminance of paper-based OLED was 3578 cd m^{-2}. The PVP film reduced the surface roughness from 71.7 nm to 5.3 nm. A smooth surface of substrate can lower the leakage current and improved current efficiency of OLED.

The hot press was alternatives to modified process of paper substrate. The dry film photoresist (DFR) had a uniform thickness and could be easily deposited on the paper substrate by hot pressing, which renders the

DFR an excellent candidate for the planarization layer of the paper-based OLED (Kim and Hwang, 2018). The turn-on voltage was 3.5 V, and the maximum luminance was 157 cd m^{-2} at 7.5 V. The polypropylene (PP) film is also a great alternative to be a buffer layer of paper substrate by hot-press method. A copy paper was successively sandwiched between two pieces of oriented polypropylene films by Wang's group (Figure 9B)(Huang et al., 2019). The composited PP paper was formed by hot pressing. And the electrode was fabricated by silver mirror reaction. Finally, an Ag-paper based OLED can be operated normally. Yang et al. also prepared the paper through a simple hot-press process with commercial paper and PP film (Figure 9C) (Yang et al., 2020). Then, the dopamine acts as an adhesion layer between paper and Cu NPs electrode. The performance of the paper-based OLED with a capping layer was characterized by a turn-on voltage of 2.5 V and a maximum luminance of 700 cd m^{-2}.

The rough surface and poor moisture resistance of commercial paper substrates greatly limit its application in OLEDs. In addition, the paper substrate usually has a low transmittance, and it is difficult to emit light through the substrate, which also greatly limits the types of OLED devices that can be prepared on paper substrates. Therefore, the transparent nanopaper composed by cellulose with non-traditional crafts is a more excellent alternative of paper-based OLED compared with conventional paper. In the past ten years, there have been relatively a lot of researches on transparent nanopaper-based OLEDs, and their efficiency is relatively high, which can be comparable to PET-based OLEDs, and the advantages of paper-based OLEDs allow it to be applied in some specific fields, such as disposable electronic devices and e-paper.

Purandare fabricated OLEDs on flexible and transparent reconstituted cellulose obtained from wood pulp (Purandare et al., 2014). The HOMO/LUMO energy levels and device structure of OLED are shown in Figure 9D. The CNC/CPI hybrid substrate with high optical, mechanical, and thermal properties was applied as OLED substrate by Tao's group (Chen et al., 2020). The nanopaper-based OLED adopted a conventional bottom-emitting OLED since the high transmittance (>85%) of the CNC/CPI hybrid substrate. The green OLED units were built on 100-μm thick pure CPI substrates, 100-μm thick 4% CNC/CPI substrates, and high-temperature-resistant glass substrates. The luminance of OLED at 7 V based on glass, CNC/CPI, and CPI were 48 347, 13 980, and 8079 cd m^{-2}, respectively. The cellulose nanofibrils (CNFs)/polyarylate (PAR) hybrid polymer substrate also was an excellent substrate with greatly improved thermal stability. Tao et al. fabricated the green OLEDs on a 120-μm thick 4 wt % CNF/PAR substrate, with a PET substrate of the same thickness as the comparison (Tao et al., 2020). The maximum luminance of CNF/PAR-based OLED and PET-based OLED were 9015 and 5142 cd m^{-2}, respectively. Moreover, CNF/PAR-based OLED obtains a higher current efficiency (CE) of 5.65 cd A^{-1} and a higher EQE of 1.68%. Zheng et al. fabricated (OLED) with enhanced stability and light extraction efficiency was prepared by using a PEDOT:PSS PH1000 transparent conductive electrode coated on regenerated cellulose film (RCF) (Zheng et al., 2021). The performances of cellulose-based OLEDs are shown in Figure 9E. The OLED with a similar structure based on ITO/RCF was also fabricated for comparison. The maximum luminance was higher than 10,000 cd m^{-2}, and the maximum EQE was 3.65%.

CHALLENGES AND CONCLUSION

In this review, we combed the development history of paper and related research on paper-based organic optoelectronic devices. The paper substrate has great application potential as a flexible substrate for organic optoelectronic devices. Compared with conventional flexible substrates such as plastic substrates, paper substrates have unique advantages, such as low cost, raw materials are abundant, good flexibility, and it can be recycled in an environmentally friendly manner. All of these make paper-based devices attractive. Cheap prices can reduce the cost of devices. The stock of cellulose, the raw material for paper, is abundant on the earth, which prevents us from being constrained by insufficient raw materials. Good flexibility makes it perfectly suitable for roll-to-roll technology, and it can be folded, which is difficult for other substrates. The characteristics of environmentally friendly recycling are precious to the earth plagued by electronic waste. The characteristics of environmental protection and recycling make paper-based electronic devices have great potential for application in disposable electronic products. The advantages of paper substrates and the results they bring are difficult to obtain with other traditional substrates.

Even though paper substrates have so many attractive advantages, the applications of paper substrates in flexible organic optoelectronic devices are still relatively few because the inherent properties of the paper substrate limit its application in optoelectronic devices. The poor surface morphology makes the paper-based organic optoelectronic device leakage current larger, and even causes the device to short-circuit.

In addition, the poor moisture and oxygen barrier properties of paper substrates make paper-based devices have lower reliability. Therefore, the paper substrate needs to be pretreated before it can be used in flexible electronic devices. At present, the effects of various methods for processing paper substrates are not satisfactory. Most research on paper-based devices uses nanopaper substrates. These paper substrates are synthesized by combining cellulose with various other materials. Organic optoelectronic devices based on nanopaper substrates have relatively good performance and high reliability. However, the complex synthesis method limits its wide range of applications. Although paper substrates have poor moisture and oxygen barrier properties, if combined with a good encapsulation treatment, paper-based organic optoelectronic devices can also exhibit excellent water and oxygen barrier properties and stability And paper can be a reliable choice for flexible substrates for organic optoelectronic devices.

Due to the special properties of paper substrates, the application of paper substrates in organic optoelectronic devices has very good prospects. However, its inherent characteristics and relatively complex synthesis methods respectively limit the application of commercial paper and nanopaper in organic optoelectronic devices. Therefore, simple and efficient substrate processing methods and simple and inexpensive nanopaper synthesis methods are very important for paper-based organic optoelectronic devices. With the advancement of scientific research and technology, we believe that the performance of paper-based organic optoelectronic devices will gradually be optimized. Therefore, we believe that paper substrates will have great potential to become the next generation of flexible substrates for organic optoelectronic devices.

ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China (Grant Nos. 62174067, 62175085, 61905086), Science and Technology Development Planning of Jilin Province (Project Nos. 20190101024JH, 20200201296JC).

AUTHOR CONTRIBUTIONS

Conceptualization, W. F. X. and T. P.; investigation, S. H. L. and T. P.; writing – original draft, T. P.; writing–review & editing, W. F. X., L.T. Z., and S. H. L.

DECLARATION OF INTERESTS

The authors declare no competing interests.

REFERENCES

Alava, M., and Niskanen, K. (2006). The physics of paper. Rep. Prog. Phys. *69*, 669–723.

Amini, E., Hafez, I., Tajvidi, M., and Bousfield, D.W. (2020). Cellulose and lignocellulose nanofibril suspensions and films: a comparison. Carbohydr. Polym. *250*, 117011.

Barsotti, J., Rapidis, A.G., Hirata, I., Greco, F., Cacialli, F., and Mattoli, V. (2021). Ultrathin, ultra-conformable, and free-standing tattooable organic light-emitting diodes. Adv. Electron. Mater. *7*, 2001145.

Brody, T.P. (1984). The thin film transistor—a late flowering bloom. IEEE Trans. Electron Devices *31*, 1614–1628.

Brooke, R., Edberg, J., Crispin, X., Berggren, M., Engquist, I., and Jonsson, M.P. (2019). Greyscale and paper electrochromic polymer displays by UV patterning. Polymers *11*, 267.

Brusso, B.C. (2021). Development of stone writing and packaging materials history. IEEE Ind. Appl. Mag. *27*, 8–11.

Burlingame, Q., Ball, M., and Loo, Y.-L. (2020). It's time to focus on organic solar cell stability. Nat. Energy *5*, 947–949.

Chaudhary, A., Poddar, M., Pathak, D.K., Misra, R., and Kumar, R. (2020). Electron donor ferrocenyl phenothiazine: counter ion for improving all-organic electrochromism. ACS Appl. Electron. Mater. *2*, 2994–3000.

Che, B., Zhou, D., Li, H., He, C., Liu, E., and Lu, X. (2019). A highly bendable transparent electrode for organic electrochromic devices. Org. Electron. *66*, 86–93.

Chen, L., Yu, H., Dirican, M., Fang, D., Tian, Y., Yan, C., Xie, J., Jia, D., Liu, H., Wang, J., et al. (2020). Highly transparent and colorless nanocellulose/polyimide substrates with enhanced thermal and mechanical properties for flexible OLED displays. Adv. Mater. Interfaces *7*, 2000928.

Costa, S.V., Pingel, P., Janietz, S., and Nogueira, A.F. (2016). Inverted organic solar cells using nanocellulose as substrate. J. Appl. Polym. Sci. *133*, 43679.

Deng, W., Xiao, Y., Lu, B., Zhang, L., Xia, Y., Zhu, C., Zhang, X., Guo, J., Zhang, X., and Jie, J. (2021). Water-surface drag coating: a new route toward high-quality conjugated small-molecule thin films with enhanced charge transport properties. Adv. Mater. *33*, 2005915.

Dov, N.E., Shankar, S., Cohen, D., Bendikov, T., Rechav, K., Shimon, L.J.W., Lahav, M., and van der Boom, M.E. (2017). Electrochromic metallo-organic nanoscale films: fabrication, color range, and devices. J. Am. Chem. Soc. *139*, 11471–11481.

Droguet, B.E., Liang, H.-L., Frka-Petesic, B., Parker, R.M., De Volder, M.F.L., Baumberg, J.J., and Vignolini, S. (2021). Large-scale fabrication of structurally coloured cellulose nanocrystal films and effect pigments. Nat. Mater. *10*, 1038.

Fang, Z., Zhu, H., Yuan, Y., Ha, D., Zhu, S., Preston, C., Chen, Q., Li, Y., Han, X., Lee, S., et al. (2014). Novel nanostructured paper with ultrahigh transparency and ultrahigh haze for solar cells. Nano Lett. *14*, 765–773.

Fujisaki, Y., Koga, H., Nakajima, Y., Nakata, M., Tsuji, H., Yamamoto, T., Kurita, T., Nogi, M., and Shimidzu, N. (2014). Transparent nanopaper-based flexible organic thin-film transistor array. Adv. Funct. Mater. *24*, 1657–1663.

Fukuda, K., Yu, K., and Someya, T. (2020). The future of flexible organic solar cells. Adv. Energy Mater. *10*, 2000765.

Gao, L., Chao, L., Hou, M., Liang, J., Chen, Y., Yu, H.-D., and Huang, W. (2019). Flexible, transparent nanocellulose paper-based perovskite solar cells. npjFlex. Electron. *3*, 4.

Ha, M.-Y., Yoon, D.-Y., Park, D.-Y., Choi, S.-J., and Moon, D.-G. (2016). Flexible green phosphorescent organic light-emitting devices on copy paper substrates. In International Workshop on Active-Matrix Flatpanel Displays and devices(AM-FPD) (IEEE), pp. 102–103.

Hall, A.K.I., Plaut, R.H., and McGuiggan, P.M. (2019). Comparing non-destructive mechanical testing methods for the assessment of brittle papers-the cantilever, hanging pear loop, and clamped fold tests. Restaurator *40*, 15–34.

Han, J.S., Jung, S.Y., Kang, D.S., and Seo, Y.B. (2020). Development of flexible calcium carbonate for papermaking filler. ACS Sustain. Chem. Eng. *8*, 8994–9001.

Hasan, I., and Walia, S. (2021). A review on properties and challenges associated with cellulose nanocrystals and nanocomposites. In International Conference on Advances in Materials Research (ICAMR), *45* (Bannari Amman Institute of Technology), pp. 3365–3369.

He, Z., Wang, L., Liu, G.-S., Xu, Y., Qu, Z., Zhong, M., Li, X., Gui, X., Lin, Y.-S., Qin, Z., et al. (2020). Constructing electrophoretic displays on foldable paper-based electrodes by a facile transferring method. ACS Appl. Electron. Mater. *2*, 1335–1342.

Hoeng, F., Denneulin, A., and Bras, J. (2016). Use of nanocellulose in printed electronics: a review. Nanoscale *8*, 13131–13154.

Hsieh, M.-C., Koga, H., Nogi, M., and Suganuma, K. (2014). Highly heat-resistant bio-based nanofiber substrate for flexible electronics. In IEEE CPMT Symposium Japan (IEEE).

Huang, J., Zhu, H., Chen, Y., Preston, C., Rohrbach, K., Cumings, J., and Hu, L. (2013). Highly transparent and flexible nanopaper transistors. ACS Nano *7*, 2106–2113.

Huang, Q., Zhang, K., Yang, Y., Ren, J., Sun, R., Huang, F., and Wang, X. (2019). Highly smooth, stable and reflective Ag-paper electrode enabled by silver mirror reaction for organic optoelectronics. Chem. Eng. J. *370*, 1048–1056.

Hubbe, M.A., and Bowden, C. (2009). Handmade paper: a review of its history, craft, and science. BioResources *4*, 1736–1792.

Hyun, W.J., Park, O.O., and Chin, B.D. (2013). Foldable graphene electronic circuits based on paper substrates. Adv. Mater. *25*, 4729–4734.

Hyun, W.J., Secor, E.B., Rojas, G.A., Hersam, M.C., Francis, L.F., and Frisbie, C.D. (2015). All-printed, foldable organic thin-film transistors on glassine paper. Adv. Mater. *27*, 7058–7064.

Isogai, A. (2021). Emerging nanocellulose technologies: recent developments. Adv. Mater. *33*, 2000630.

Jain, P., and Gupta, C. (2021). A sustainable journey of handmade paper from past to present: a review. Probl. Ekorozwoju *16*, 234–244.

Jayaraman, E., and Iyer, S.S.K. (2020). Organic photovoltaic modules built on paper substrates. Adv. Mater. Technol. *5*, 2000664.

Ji, Y., Xu, Q., Jin, L., and Fu, Y. (2020). Cellulosic paper with high antioxidative and barrier properties obtained through incorporation of tannin into kraft pulp fibers. Int. J. Biol. Macromol. *162*, 678–684.

Jiang, Y., Wang, Z., Liu, X., Yang, Q., Huang, Q., Wang, L., Dai, Y., Qin, C., and Wang, S. (2020). Highly transparent, UV-shielding, and water-resistant lignocellulose nanopaper from agro-industrial waste for green optoelectronics. ACS Sustain. Chem. Eng. *8*, 17508–17519.

Jin, J., Lee, D., Im, H.-G., Han, Y.C., Jeong, E.G., Rolandi, M., Choi, K.C., and Bae, B.-S. (2016). Chitin nanofiber transparent paper for flexible green electronics. Adv. Mater. *28*, 5169–5175.

Kang, K.S., Jeong, S.Y., Jeong, E.G., and Choi, K.C. (2020). Reliable high temperature, high humidity flexible thin film encapsulation using Al_2O_3/MgO nanolaminates for flexible OLEDs. Nano Res. *13*, 2716–2725.

Kim, S., and Hwang, B. (2018). Ag nanowire electrode with patterned dry film photoresist insulator for flexible organic light-emitting diode with various designs. Mater.Des. *160*, 572–577.

Kim, Y.M., Li, X., Kim, K.-W., Kim, S.H., and Moon, H.C. (2019). Tetrathiafulvalene: effective organic anodic materials for WO3-based electrochromic devices. RSC Adv. *9*, 19450–19456.

Kim, D., Reddy, M.R., Cho, K., Ho, D., Kim, C., and Seo, S. (2020). Solution-processable fluorene derivative for organic thin-film transistors. Org. Electron. *76*, 105464.

Kim, S.W., Park, S., Lee, S., Kim, D., Lee, G., Son, J., and Cho, K. (2021). Stretchable mesh-patterned organic semiconducting thin films on creased elastomeric substrates. Adv. Funct. Mater. *31*, 2010870.

Kraft, U., Zaki, T., Letzkus, F., Burghartz, J.N., Weber, E., Murmann, B., and Klauk, H. (2019). Low-voltage, high-frequency organic transistors and unipolar and complementary ring oscillators on paper. Adv. Electron. Mater. *5*, 1800453.

Kroeling, H., Duchemin, B., Dormanns, J., Schabel, S., and Staiger, M.P. (2018). Mechanical anisotropy of paper-based all-cellulose composites. Compos.AAppl. Sci. Manuf. *113*, 150–157.

Lee, B.-S., Park, B., Yang, H.-S., Han, J.W., Choong, C., Bae, J., Lee, K., Yu, W.-R., Jeong, U., Chung, U.I., et al. (2014). Effects of substrate on piezoelectricity of electrospun poly(vinylidene fluoride)-nanofiber-based energy generators. ACS Appl. Mater. Interfaces *6*, 3520–3527.

Lee, H.B., Jin, W.-Y., Ovhal, M.M., Kumar, N., and Kang, J.-W. (2019). Flexible transparent conducting electrodes based on metal meshes for organic optoelectronic device applications: a review. J. Mater. Chem. C *7*, 1087–1110.

Li, Y., Liu, C., Xu, Y., Minari, T., Darmawan, P., and Tsukagoshi, K. (2012). Solution-processed organic crystals for field-effect transistor arrays with smooth semiconductor/dielectric interface on paper substrates. Org. Electron. *13*, 815–819.

Li, H., Li, X., Wang, W., Huang, J., Li, J., Lu, Y., Chang, J., Fang, J., and Song, W. (2019). Highly foldable and efficient paper-based perovskite solar cells. Sol. RRL *3*, 1800317.

Li, T., Chen, Z., Wang, Y., Tu, J., Deng, X., Li, Q., and Li, Z. (2020). Materials for interfaces in organic solar cells and photodetectors. ACS Appl. Mater. Interfaces *12*, 3301–3326.

Lim, K.-G., Han, T.-H., and Lee, T.-W. (2021). Engineering electrodes and metal halide perovskite materials for flexible/stretchable perovskite solar cells and light-emitting diodes. Energy Environ. Sci. *14*, 2009–2035.

Liu, Y., and Chen, Y. (2020). Integrated perovskite/bulk-heterojunction organic solar cells. Adv. Mater. *32*, 1805843.

Liu, G., Liu, Y., Zhang, M., Pettersson, F., and Toivakka, M. (2020a). Fabrication of all-solid organic electrochromic devices on absorptive paper substrates utilizing a simplified lateral architecture. Materials *13*, 4839.

Liu, Y., Hu, C., Wu, J., Islam, A., and Ouyang, X. (2020b). Carboxy cellulose as cathode interfacial layer for efficient organic solar cells. Paper Biomater. *5*, 14–21.

Liu, S., Zang, C., Zhang, J., Tian, S., Wu, Y., Shen, D., Zhang, L., Xie, W., and Lee, C.-S. (2022). Air-stable ultrabright inverted organic light-emitting devices with metal ion-chelated polymer injection layer. Nanomicro Lett. *14*, 14.

Lu, Z., Wang, C., Deng, W., Achille, M.T., Jie, J., and Zhang, X. (2020). Meniscus-guided coating of organic crystalline thin films for high-performance organic field-effect transistors. J. Mater. Chem. C *8*, 9133–9146.

Makita, T., Ninomiya, Y., Kumagai, S., Okamoto, T., Sasaki, M., Watanabe, S., and Takeya, J. (2021). Nano-ground glass as a superhydrophilic template for printing high-performance organic single-crystal thin films. Adv. Mater. Interfaces *8*, 2100033.

Nogi, M., Kim, C., Sugahara, T., Inui, T., Takahashi, T., and Suganuma, K. (2013). High thermal stability of optical transparency in cellulose nanofiber paper. Appl. Phys. Lett. *102*, 181911.

Park, J., Seo, J.-H., Yeom, S.-W., Yao, C., Yang, V.W., Cai, Z., Jhon, Y.M., and Ju, B.-K. (2018). Flexible and transparent organic phototransistors on biodegradable cellulose nanofibrillated fiber substrates. Adv. Opt. Mater. *6*, 1701140.

Peng, B., and Chan, P.K.L. (2014). Flexible organic transistors on standard printing paper and memory properties induced by floated gate electrode. Org. Electron. *15*, 203–210.

Peng, X., Liu, H., Chen, P., Tang, F., Hu, Y., Wang, F., Pi, Z., Zhao, M., Chen, N., Chen, H., et al. (2019). A chromosome-scale genome assembly of paper mulberry (Broussonetia papyrifera) provides new insights into its forage and papermaking usage. Mol. Plant *12*, 661–677.

Pinto, E.R.P., Barud, H.S., Silva, R.R., Palmieri, M., Polito, W.L., Calil, V.L., Cremona, M., Ribeiro, S.J.L., and Messaddeq, Y. (2015). Transparent composites prepared from bacterial cellulose and castor oil based polyurethane as substrates for flexible OLEDs. J. Mater. Chem. C *3*, 11581–11588.

Purandare, S., Gomez, E.F., and Steckl, A.J. (2014). High brightness phosphorescent organic light emitting diodes on transparent and flexible cellulose films. Nanotechnology *25*, 094012.

Qian, C., Sun, J., Yang, J., and Gao, Y. (2015). Flexible organic field-effect transistors on biodegradable cellulose paper with efficient reusable ion gel dielectrics. RSC Adv. *5*, 14567–14574.

Raghuwanshi, V., Bharti, D., Mahato, A.K., Varun, I., and Tiwari, S.P. (2019). Solution-processed organic field-effect transistors with high performance and stability on paper substrates. ACS Appl. Mater. Interfaces *11*, 8357–8364.

Rahman, M.M., and Rimu, S.H. (2020). Recent development in cellulose nanocrystal-based hydrogel for decolouration of methylene blue from aqueous solution: a review. Int. J. Environ.Anal.Chem. *10*, 1080.

Rawat, M., Jayaraman, E., Balasubramanian, S., and Iyer, S.S.K. (2019). Organic solar cells on paper substrates. Adv. Mater. Technol. *4*, 1900184.

Reimer, M., and Zollfrank, C. (2021). Cellulose for light manipulation: methods, applications, and prospects. Adv. Energy Mater. *11*, 2003866.

Robinson, A. (2013). Technology: pulp, pilcrows and interrobangs. Nature *501*, 491.

Sarrazin, P., Valecce, L., Beneventi, D., Chaussy, D., Vurth, L., and Stephan, O. (2007). Photoluminescent paper based on poly (fluorene-co-fluorenone) particles adsorption on modified cellulose fibers. Adv. Mater. *19*, 3291–3294.

Solala, I., Iglesias, M.C., and Peresin, M.S. (2020). On the potential of lignin-containing cellulose nanofibrils (LCNFs): a review on properties and applications. Cellulose *27*, 1853–1877.

Son, S.-Y., Yang, S.-L., Lee, S., Na, S.-I., and Joh, H.-I. (2020). Eco-friendly cellulose-derived transparent carbon nanosheet electrodes. Mater. Res. Bull. *132*, 110999.

Song, Z., Xiao, H., and Zhao, Y. (2014). Hydrophobic-modified nano-cellulose fiber/PLA biodegradable composites for lowering water vapor transmission rate (WVTR) of paper. Carbohydr.Polym. *111*, 442–448.

Song, J.-K., Kim, M.S., Yoo, S., Koo, J.H., and Kim, D.-H. (2021). Materials and devices for flexible and stretchable photodetectors and light-emitting diodes. Nano Res. *14*, 2919–2937.

Sonmez, G., Meng, H., and Wudl, F. (2004). Organic polymeric electrochromic devices: polychromism with very high coloration efficiency. Chem. Mater. *16*, 574–580.

Sun, Y., Chang, M., Meng, L., Wan, X., Gao, H., Zhang, Y., Zhao, K., Sun, Z., Li, C., Liu, S., et al. (2019). Flexible organic photovoltaics based on water-processed silver nanowire electrodes. Nat. Electron. *2*, 513–520.

Tao, J., Wang, R., Yu, H., Chen, L., Fang, D., Tian, Y., Xie, J., Jia, D., Liu, H., Wang, J., et al. (2020). Highly transparent, highly thermally stable nanocellulose/polymer hybrid substrates for flexible OLED devices. ACS Appl. Mater. Interfaces *12*, 9701–9709.

Tobjork, D., and Osterbacka, R. (2011). Paper electronics. Adv. Mater. *23*, 1935–1961.

Tu, H., Zhu, M., Duan, B., and Zhang, L. (2021). Recent progress in high-strength and robust regenerated cellulose materials. Adv. Mater. *33*, 2000682.

Ummartyotin, S., Juntaro, J., Sain, M., and Manuspiya, H. (2012). Development of transparent bacterial cellulose nanocomposite film as substrate for flexible organic light emitting diode (OLED) display. Ind. Crop Prod. *35*, 92–97.

Voggu, V.R., Sham, J., Pfeffer, S., Pate, J., Fillip, L., Harvey, T.B., Brown, R.M., Jr., and Korgel, B.A. (2017). Flexible CuInSe2 nanocrystal solar cells on paper. ACS Energy Lett. *2*, 574–581.

Wan, J., Zhang, L., He, Q., Liu, S., Huang, B., Hu, L., Zhou, W., and Chen, Y. (2020). High-performance pseudoplanar heterojunction ternary organic solar cells with nonfullerene alloyed acceptor. Adv. Funct. Mater. *30*, 1909760.

Wang, C.-Y., Fuentes-Hernandez, C., Liu, J.-C., Dindar, A., Choi, S., Youngblood, J.P., Moon, R.J., and Kippelen, B. (2015). Stable low-voltage operation top-gate organic field-effect transistors on cellulose nanocrystal substrates. ACS Appl. Mater. Interfaces *7*, 4804–4808.

Wang, C.-Y., Fuentes-Hernandez, C., Chou, W.-F., and Kippelen, B. (2017). Top-gate organic field-effect transistors fabricated on paper with high operational stability. Org. Electron. *41*, 340–344.

Wang, H., Zang, C., Shan, G., Yu, Z., Liu, S., Zhang, L., Xie, W., and Zhao, H. (2019). Bluish-green thermally activated delayed fluorescence material for blue-hazard free hybrid white organic light-emitting device with high color quality and low efficiency roll-off. Adv. Opt. Mater. *7*, 1801718.

Wang, Y., Liu, X., Sheng, J., Zhu, H., and Yang, R. (2021). Nanoporous regenerated cellulose separator for high-performance lithium ion batteries prepared by nonsolvent-induced phase separation. ACS Sustain. Chem. Eng. *9*, 14756–14765.

Weng, G., Yang, Y., Zhao, J., Li, J., Zhu, J., and Zhao, J. (2020). Improving the SERS enhancement and reproducibility of inkjet-printed Au NP paper substrates by second growth of Ag nanoparticles. Mater. Chem. Phys. *253*, 123416.

Wu, J., Che, X., Hu, H.-C., Xu, H., Li, B., Liu, Y., Li, J., Ni, Y., Zhang, X., and Ouyang, X. (2020a). Organic solar cells based on cellulose nanopaper from agroforestry residues with an efficiency of over 16% and effectively wide-angle light capturing. J. Mater. Chem.A *8*, 5442–5448.

Wu, J., Liu, Y., Islam, A., Zheng, Q., Li, J., Ji, W., Chen, L., and Ouyang, X. (2020b). From straw to device interface: carboxymethyl-cellulose-based modified interlayer for enhanced power conversion efficiency of organic solar cells. Adv. Sci. *7*, 1902269.

Xu, M., Zhang, X., Li, S., Xu, T., Xie, W., and Wang, W. (2019). Gate-controlled multi-bit nonvolatile ferroelectric organic transistor memory on paper substrates. J. Mater. Chem. C *7*, 13477–13485.

Xu, Y., Fei, Q., Page, M., Zhao, G., Ling, Y., Stoll, S.B., and Yan, Z. (2021). Paper-based wearable electronics. iScience *24*, 102736.

Xue, Q., and Xie, G. (2021). Thermally activated delayed fluorescence beyond through-bond charge transfer for high-performance OLEDs. Adv. Opt. Mater. *9*, 2002204.

Yang, Y., Liu, W., Huang, Q., Li, X., Ling, H., Ren, J., Sun, R., Zou, J., and Wang, X. (2020). Full solution-processed fabrication of conductive hybrid paper electrodes for organic optoelectronics. ACS Sustain. Chem. Eng. *8*, 3392–3400.

Yao, Y., Tao, J., Zou, J., Zhang, B., Li, T., Dai, J., Zhu, M., Wang, S., Fu, K.K., Henderson, D., et al. (2016). Light management in plastic-paper hybrid substrate towards high-performance optoelectronics. Energy Environ. Sci. *9*, 2278–2285.

Yoon, D.-Y., and Moon, D.-G. (2012). Bright flexible organic light-emitting devices on copy paper substrates. Curr.Appl. Phys. *12*, E29–E32.

Yoon, D.-Y., Kim, T.-Y., and Moon, D.-G. (2010). Flexible top emission organic light-emitting devices using sputter-deposited Ni films on copy paper substrates. Curr. Appl. Phys. *10*, E135–E138.

Yu, Z., Zhang, J., Liu, S., Zhang, L., Zhao, Y., Zhao, H., and Xie, W. (2019). High-efficiency blue phosphorescent organic light-emitting devices with low efficiency roll-off at ultrahigh luminance by the reduction of triplet-polaron quenching. ACS Appl. Mater. Interfaces *11*, 6292–6301.

Zakaria, S., Ong, B., Ahmad, S., Abdullah, M., and Yamauchi, T. (2005). Preparation of lumen-loaded kenaf pulp with magnetite (Fe_3O_4). Mater. Chem. Phys. *89*, 216–220.

Zang, C., Liu, S., Xu, M., Wang, R., Cao, C., Zhu, Z., Zhang, J., Wang, H., Zhang, L., Xie, W., et al. (2021). Top-emitting thermally activated delayed fluorescence organic light-emitting devices with weak light-matter coupling. LightSci. Appl. *10*, 116.

Zhang, X., Liu, S., Zhang, L., and Xie, W. (2019a). In-planar-electrodes organic light-emitting devices for smart lighting applications. Adv. Opt. Mater. *7*, 1800857.

Zhang, X., Pan, T., Zhang, J., Zhang, L., Liu, S., and Xie, W. (2019b). Color-tunable, spectra-stable flexible white top-emitting organic light-emitting devices based on alternating current driven and dual-microcavity technology. ACS Photonics *6*, 2350–2357.

Zhao, S.-Q., Jiang, Z.-Y., Chen, L.-S., Huang, W., and Liu, Y.-H. (2021). Self-supporting, ultra-thin and highly transparent conducting nickel grids for extremely flexible and stretchable electrochromic devices. Opt. Express *29*, 25254–25269.

Zheng, X., Huang, R., Zhong, C., Xie, G., Ning, W., Huang, M., Ni, F., Dias, F., and Yang, C. (2020). Achieving 21% external quantum efficiency for nondoped solution-processed sky-blue thermally activated delayed fluorescence OLEDs by means of multi-(donor/acceptor) emitter with throughspace/-bond charge transfer. Adv. Sci. *7*, 1902087.

Zheng, Q., Li, H., Zheng, Y., Li, Y., Liu, X., Nie, S., Ouyang, X., Chen, L., and Ni, Y. (2021). Cellulose-based flexible organic light-emitting diodes with enhanced stability and external quantum efficiency. J. Mater. Chem. C *9*, 4496–4504.

Zhong, Z., Peng, X., and Iop. (2019). Study on the manufacturing technology of Guangdong hand-made paper. In 5th International Conference on Advances in Energy, Environment and Chemical Engineering (AEECE), *358* (Beijing University of Agriculture, College of Resources and Environmental Sciences), p. 042064, Paper presented at.

Zhou, Y., Khan, T., Liu, J., Fuentes-Hernandez, C., Shim, J., Najafabadi, E., Youngblood, J., Moon, R., and Kippelen, B. (2014). Efficient recyclable organic solar cells on cellulose nanocrystal substrates with a conducting polymer top electrode deposited by film-transfer lamination. Org. Electron. *15*, 661–666.

Zhu, H., Jia, Z., Chen, Y., Weadock, N., Wan, J., Vaaland, O., Han, X., Li, T., and Hu, L. (2013a). Tin anode for sodium-ion batteries using natural wood fiber as a mechanical buffer and electrolyte reservoir. Nano Lett. *13*, 3093–3100.

Zhu, H., Xiao, Z., Liu, D., Li, Y., Weadock, N.J., Fang, Z., Huang, J., and Hu, L. (2013b). Biodegradable transparent substrates for flexible organic-light-emitting diodes. Energy Environ. Sci. *6*, 2105–2111.

Zhu, M., Cao, J., Wei, X., He, Y., Li, A., Xu, X., Ali, M.U., Yan, L., and Meng, H. (2018). Self-supported hysteresis-free flexible organic thermal transistor based on commercial graphite paper. Appl. Phys. Lett. *112*, 253301.

Zocco, A.T., You, H., Hagen, J.A., and Steckl, A.J. (2014). Pentacene organic thin-film transistors on flexible paper and glass substrates. Nanotechnology *25*, 094005.