Article type: Review

Paper in Electronic and Optoelectronic Devices

Dongheon Ha,* Zhiqiang Fang, and Nikolai B. Zhitenev

Dr. Dongheon Ha, Dr. Nikolai B. Zhitenev
Center for Nanoscale Science and Technology, National Institute of Standards and Technology, Gaithersburg, Maryland, 20899, USA
E-mail: dongheon.ha@nist.gov

Dr. Dongheon Ha
Maryland Nanocenter, University of Maryland, College Park, Maryland, 20742, USA

Prof. Zhiqiang Fang
State Key Laboratory of Pulp and Paper Engineering, South China University of Technology, Guangzhou, 510640, China

Prof. Zhiqiang Fang
South China Institute of Collaborative Innovation, South China University of Technology, Dongguan, 523808, China

Keywords: paper, electronics, optoelectronics, flexible, cellulose

Abstract

Paper, one of the oldest materials for storage and exchange of human’s information, has been reinvented as a building component of electronic and optoelectronic devices over the past decades with successful demonstration of paper-based or paper-using devices. These recent achievements can meet the demand for lightweight, cost-effective, and/or flexible electronic and optoelectronic devices with advanced functionality and reduced manufacturing costs. This article provides a review of electronic and optoelectronic devices relying on or making use of the unique properties achievable with paper-based materials. Basic scientific/technical principles, quantitative comparisons of material, electronic and/or optical properties, and benefits for each paper-based application are given. Application-specific research challenges, future design considerations, and development directions are also discussed.
1. Introduction

Since the invention of papermaking technique around AD 105,[1] paper has been one of the most widely used materials for storage and exchange of information in everyday life. At present, we use paper for a variety of purposes, such as writing, drawing, packing, etc. Typically, paper is comprised of cellulose fibers with diameters of 10 μm to 50 μm and lengths of several millimeters.[2] The cell wall of a cellulose fiber consists of a bunch of microfibrils (5 nm to 50 nm in diameter and several microns in length). The microfibrils themselves are composed of self-assembled elementary fibrils, with a diameter of ≈ 3.5 nm.[3-5]

Recently, the possibility of paper applications in electronic and optoelectronic devices has attracted a lot of attention due to its cost-effectiveness, ease of mass production, flexibility, natural abundance, and environmental-friendliness.[6-8] The price of paper (≈ 10 ¢/m²), which is much lower than that of conventional plastics such as polyethylene terephthalate (PET) (≈ 2 $/m²),[9] its flexibility, and compatibility with high speed roll-to-roll processes, all combine to make regular paper one of the strongest candidates to replace more conventional plastic substrates for electronics. However, several disadvantages of cellulosic paper such as microscale surface roughness, opaqueness, and high porosity have delayed its incorporation into industrial applications.

To mitigate some of these obstacles, many researchers are fabricating new types of paper with reduced surface roughness and/or increasing transparency.[7, 10-29] To overcome high surface roughness, various methods have been explored including patterning devices on other flat substrates that can later be attached to paper[30, 31] or coating another smooth layer on the paper surface.[32-35] Also, papers based on microfibrils or nanofibrillated cellulose (NFC) are being developed, which provide smoother substrates as compared to regular paper. To increase transparency, special papermaking techniques, such as a chemical pretreatment of cellulose fibers (e.g., 2,2,6,6-Tetramethylpiperidine-1-oxyl radical (TEMPO)),[11, 36-40] the use
of cellulose nanofibers as starting materials,\textsuperscript{[41]} or the infiltration of pores within paper with a transparent material,\textsuperscript{[9, 42-44]} have been applied.

Schematics in Fig. 1a-c illustrate how transparent paper is made with the TEMPO oxidation. The TEMPO oxidation undermines the hydrogen bonds between the cellulose fibrils, forcing the wood fibers to swell up and then collapse during papermaking process. Scanning electron microscope (SEM) images in Fig. 1d show microscale cellulosic structure of a regular paper and of a transparent paper made of TEMPO-oxidized wood fibers. The transparent paper has a higher packing density of cellulose fibers as well as fewer and smaller porous cavities than a regular paper. This reduces the backward scattering of incident light within the layer leading to high transmission.

For electronic applications, electrical properties such as dielectric constant and resistivity are of particular importance. These properties are determined or strongly influenced by the paper microstructure. For example, the dielectric constant of dry cellulose is around 6 to 8, but the porous structure of paper lowers its dielectric constant to values of around 1.3 to 4 at 1 MHz.\textsuperscript{[9]} To be used as a gate insulator in electronic devices, a higher dielectric constant is required and impregnating the porous structure with materials having higher dielectric constants (\textit{e.g.}, oils, resins, or electrolytes) is necessary.\textsuperscript{[45]} Some external factors, such as relative humidity, moisture, temperature, frequency, etc.,\textsuperscript{[46-49]} significantly impact these properties. It is recently revealed that, for chiral cellulose nanocrystals (CNCs) films, the amount of dielectric constant change with humidity is further strongly influenced by the structure and surface functionality.\textsuperscript{[50]} The conductivity of cellulose is determined by the mobility of either the lattice ions or impurity ions, and the volume resistivity is typically very large at room temperature,\textsuperscript{[51]} up to from $10^{10}$ $\Omega\cdot$cm to $10^{14}$ $\Omega\cdot$cm when the relative humidity is in the range from 20\% to 40\%.\textsuperscript{[9]} However, the resistivity of cellulose decreases to around $10^4$ $\Omega\cdot$cm when the relative humidity increases to 99\%.\textsuperscript{[51]} When paper is used as a flexible electrode with conductive materials on top, smooth and compact surface is required to reduce
resistivity. Such paper coated with thin indium zinc oxide (IZO) shows a resistivity of $3.42 \times 10^{-4} \ \Omega \cdot \text{cm}$ and a Hall mobility of $28.9 \ \text{cm}^2/\text{V} \cdot \text{s}$, confirming its potential to be utilized in electronic devices. More generally, the electrical properties of paper are correlated to a variety of environmental factors and should be carefully considered for practical electronic applications.

The optical properties of paper are equally critical for its use in optoelectronics. Regular paper is opaque, as it reflects incident light very well due to its numerous porous air gaps among cellulose fibers (see Fig. 1). This has impeded the usage of regular paper in applications that require high transmittance, such as transparent conductive electrodes, antireflection coatings (ARCs) of photovoltaics, displays, etc. Results from the aforementioned TEMPO oxidation process for highly transparent paper are promising. Papers made by this method show transmittance of $\approx 90 \%$, which is still less than that of a glass slide but is sufficient for actual application in some optoelectronic devices. One recent study also shows transparent nanocellulose paper made of cotton-based nanofibers. Densely packed nanofibers enable superior optical transmittance (more than $80 \%$ at visible spectrum, $\approx 90 \%$ at near-infrared range), which is relevant for a range of applications including paper displays, smart packaging, etc.

Another important optical property is optical haze. Depending on applications, higher or lower optical haze is required. For photovoltaic applications, such as antireflection coatings, transparent paper with high optical haze enhances the path length within solar cell absorbing materials leading to increased absorption. On the other hand, a lower optical haze is needed for indoor display panel applications to achieve higher visibility. To reduce optical haze, the porous cavities within the transparent paper can be filled with polymers, such as resins, polyvinyl alcohol (PVA), etc. Alternatively, downsizing the size of the fibers from microscale to nanoscale can be considered. However, to increase the out-coupling efficiency of organic light-emitting diodes (OLEDs), a substrate with both high
transparency and high optical haze is required if the light is emitted through the substrate.\cite{42} Thus, it is important to optimize the transmittance and the haze through tuning the porosity or the size of fibers depending on the different optoelectronic applications.

In this review, we describe some recent advances of paper usage in various electronic and optoelectronic devices and applications. Table 1 briefly summarizes paper-based applications covered in this review with some specific information about relevant electronic and/or optical properties. Research challenges and the outlook for future developments are also discussed.

Table 1. Applications of paper and the corresponding electronic and/or optical characteristics.

<table>
<thead>
<tr>
<th>Applications</th>
<th>Possible use of paper</th>
<th>Brief description of representative research</th>
</tr>
</thead>
<tbody>
<tr>
<td>Photovoltaics</td>
<td>Antireflection coatings\cite{53, 54} or substrates\cite{36, 57-65}</td>
<td>Micro-sized cellulose paper as an antireflection coating for photovoltaics: increased absorption due to index contrast and surface texturing when a transparent paper is applied atop a high index semiconductor (e.g., silicon (Si), gallium arsenide (GaAs)) or a low index polymer substrate, ≈ 24 % enhancements in the power conversion efficiency (η) of a GaAs solar cell\cite{53, 54} Cellulose nanocrystals-based paper as a substrate for an organic solar cell: short-circuit current density (J\text{sc}) of 16.1 mA/cm², open circuit voltage (V\text{oc}) of 0.68 V, fill factor (FF) of 54 %, and power conversion efficiency of 2.7 %\cite{59} Hydrophilic mesoporous material coated ultrasmooth cellulose paper as a substrate for a flexible indoor solar cell: short-circuit current density of 10.19 mA/cm², open circuit voltage of 0.82 V, fill factor of 40.7 %, and power conversion efficiency of 3.4 %\cite{65}</td>
</tr>
<tr>
<td>Electronic circuits</td>
<td>Substrate,\cite{41, 66-93} gate dielectric of field effect transistors (FETs),\cite{41, 92-97} tunnel barrier of memory devices,\cite{98, 99} or electrolyte\cite{100}</td>
<td>Nanofibrillated cellulose (NFC)-based nanopaper as a substrate of an organic FET: effective carrier mobility of $4.3 \times 10^{-3}$ cm²/V·s, drain-source current on/off modulation ratio of 200\cite{77} Ultrasmooth and clear nanopaper made of NFC acts as a substrate for In–Ga–ZnO/Al₂O₃ thin-film transistors (TFTs): saturation mobility of 15.8 cm²/V·s, on/off modulation ratio of $4.4 \times 10^5$, threshold voltage of -0.42 V, and a subthreshold gate</td>
</tr>
<tr>
<td><strong>Transparent conductive electrodes</strong></td>
<td><strong>Electrodes</strong>[^14, 56, 61, 101-121]</td>
<td>Paper as a gate dielectric layer of a flexible FET: effective carrier mobility of &gt; 30 cm²/V·s, drain-source current on/off modulation ratio of 10⁴, a near-zero threshold voltage, and a subthreshold gate voltage swing of 0.8 V per decade[^94]</td>
</tr>
<tr>
<td>-------------------------------------</td>
<td>---------------------------------</td>
<td>---</td>
</tr>
<tr>
<td><strong>Display</strong></td>
<td><strong>Substrates for OLEDs[^42, 122-131] or touch screens[^65, 66]</strong></td>
<td>Silver nanowires-coated cellulose nanofibers as a transparent conductive electrode for light-emitting diodes or solar cells: high optical transmission of &gt; 90 % and good conductivity while maintaining stable operation under bending[^118]</td>
</tr>
<tr>
<td><strong>Energy storage</strong></td>
<td><strong>Electrodes[^108-114, 116, 132-147], separators[^101, 132, 134, 148, 149], reservoir[^150, 151] or substrate[^152]</strong></td>
<td>Metal nanotrough networks coated on paper: high optical transmission of &gt; 90 % and low sheet resistance of ≈ 2 Ω per square, superior optical transmittance in near-infrared (NIR) range where conventional transparent conductive electrodes (e.g., indium tin oxide) are opaque[^103]</td>
</tr>
<tr>
<td><strong>Antennas</strong></td>
<td><strong>Substrates[^153-166]</strong></td>
<td>Transparent nanopaper-based substrate for a highly flexible OLED: strong mechanical properties with a maximum loading stress of 200 MPa to 400 MPa, low coefficient of thermal expansion (2.7 µm/m/K), high optical transmittance (&gt; 93 %) at a wavelength of 550 nm[^126]</td>
</tr>
</tbody>
</table>

[^121]: Transparent conductive electrodes.

[^122]: Displays.

[^123]: Flexible cellulose nanocomposites as substrates for an OLED: low coefficient of thermal expansion, high optical transmittance (> 82 %) in the visible spectrum.

[^125]: Transparent nanopaper-based substrate for a highly flexible OLED: high optical transmittance (> 85 %) and haze of > 90 %, more than 35.1 % improved power efficiency compared to an OLED on plastic or glass substrate.

[^126]: Plastic-paper hybrid structure as a substrate for an OLED: high optical transmittance of > 85 % and haze of > 90 %, more than 35.1 % improved power efficiency compared to an OLED on plastic or glass substrate.

[^127]: Energy storage.

[^128]: Filter paper as an electrode of a supercapacitor: an areal capacitance of 700 mF/cm² at a scan rate of 5 mV/s with maintaining capacity more than 85 % over 1000 cycles (at a current density of 20 mA/cm²).

[^129]: Conductive nanopaper as counter/reference electrodes of a lithium (Li)-ion battery: 1200 mA·h/g for 100 cycles.

[^130]: Antennas.

[^131]: 30 nm cellulose nanofibers-based paper as a substrate of a foldable antenna: return losses of -26.7 dB on compressed pulp paper, transmission and reception of multiple frequencies by folding the paper-based antenna.

[^132]: A nanopaper composite as a substrate of a flexible antenna: a minimum return loss at 2.6 GHz for Wi-Fi communication.
2. Photovoltaics

Renewable energy sources including solar energy have been increasingly important for economic and environmental reasons. Finding ways to develop high-efficiency and low-cost devices has been one of the primary goals of the solar energy industry. At a typical non-optimized interface between air and a solar cell semiconductor absorbing layer, more than 30% of the incident light is lost to reflection. Consequently, reducing such reflection is a key design principle for making high-efficiency solar cells. For this purpose, many methods to reduce reflection using dielectric thin-films,\cite{167} nanostructures,\cite{168} or plasmonics\cite{169} have been proposed. However, due to wavelength or angle dependent behavior and/or because of additional manufacturing costs, these methods have been found to be of limited practical value. Research towards more efficient and less expensive antireflection coatings remains highly relevant.

2.1. Advanced light management using paper-based antireflection coatings

Recently, novel antireflection coatings, offering attractive properties and values, have been developed using micro-sized cellulose fibers.\cite{53, 54} To make paper-based antireflection coatings, micro-sized cellulose fibers were treated with TEMPO oxidation, which results in higher packing of cellulose fibers and fewer air cavities within the layer as confirmed with SEM images in Fig. 1. The fibers used in this work have an average length of 0.8 mm, width of 27 μm, and thickness of 40 μm to 50 μm. As incident light experiences less scattering within the paper due to the improved material homogeneity, high light transmission (≈ 90%) is achieved, in contrast with regular paper that is white in color due to severe light scattering. Due to its good optical properties, this transparent paper has been tested as an antireflection coating applied atop solar cells. The transfer process to a GaAs solar cell is shown in Figure 2. This antireflection coating made of renewable and earth abundant material can be attached to solar cells without any complicated, high-temperature, or expensive vacuum-based processes,
which are necessary for conventional thin-film based antireflection coatings (e.g., thin-films made of silicon nitride (Si₃N₄), silicon dioxide (SiO₂), etc.). Polyvinyl acetate (PVAc) is used as a binding material between the paper and the GaAs substrate. The refractive index \((n)\) of PVAc \(n \approx 1.4665\) is comparable to that of cellulose \(n \approx 1.47\) and the binding material does not cause any index mismatch relative to the paper-based ARC. If required, the paper coating can also be removed from the surface without damage of the surface (Fig. 2d). As shown in Fig. 2e, randomly oriented micro-sized cellulose fibers are observed at the surface of the ARC. The roughened surface enables incident light to scatter in a wide solid angle in the ARC, resulting in path length enhancement and increased absorption, when coupled to the active material (i.e., GaAs).

To quantify the effect of this paper-based ARC, optical and electronic properties have been determined (Fig. 3). Reflectivity was measured using an integrating sphere with a monochromatic light, and analytical calculations were made accounting for multiple reflections within the layer. Both measurements and calculations show that reflectivity is reduced for a broad range of wavelengths \((\lambda = 400 \text{ nm to } 900 \text{ nm})\) and incident angles \((\theta = 10^\circ \text{ to } 55^\circ)\) due to the gradual index change within the ARC from the value of air to that of the active material (Fig. 3a–d). Furthermore, incoherent scattering associated with the textured cellulose material brings the angle-insensitive behavior, which eliminates the need for costly solar trackers. The external quantum efficiency (EQE), a ratio of the number of photo-generated electron-hole pairs to the number of incident photons, was measured with and without the paper-based ARC atop a GaAs solar cell. With the ARC, improved EQE is observed over the entire operational spectrum of a GaAs solar cell due to the reduced reflectivity (Fig. 3e). This leads to a roughly \(20\%\) increase in the short-circuit current density (Fig. 3f). Overall, the significant increase in absorptivity results in \(\approx 24\%\) enhancement of the power conversion efficiency \((\eta)\) on a planar GaAs solar cell. Similar photovoltaic property enhancements are observed on a textured Si cell as summarized in Table 2. To evaluate its
robustness under realistic operational environments, the paper-based ARC has been tested for a time equivalent to 21 sunny days (assuming 8 h of sun illumination per day, exposed to 168 h total under one sun air mass 1.5 global (AM1.5G) illumination), and no significant degradation of light transmission is observed.

Table 2. Summarized electrical properties with and without the transparent paper-based ARC on a planar GaAs\textsuperscript{[53]} and a textured Si\textsuperscript{[54]} solar cell. The uncertainties are the standard deviation of the mean.

<table>
<thead>
<tr>
<th></th>
<th>(V_{OC}) (mV)</th>
<th>(J_{SC}) (mA/cm(^2))</th>
<th>FF (%)</th>
<th>(\eta) (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bare planar GaAs solar cell</td>
<td>1001.9 ± 0.3</td>
<td>18.67 ± 0.01</td>
<td>72.5 ± 0.5</td>
<td>13.55 ± 0.10</td>
</tr>
<tr>
<td>Planar GaAs solar cell with paper-based ARC</td>
<td>1004.0 ± 0.5</td>
<td>22.49 ± 0.01</td>
<td>74.4 ± 0.2</td>
<td>16.79 ± 0.03</td>
</tr>
<tr>
<td>Enhancement</td>
<td>0.21 %</td>
<td>20.46 %</td>
<td>2.62 %</td>
<td>23.91 %</td>
</tr>
<tr>
<td>Bare textured Si solar cell</td>
<td>563.9 ± 0.3</td>
<td>28.02 ± 0.02</td>
<td>57.3 ± 1.0</td>
<td>9.05 ± 0.20</td>
</tr>
<tr>
<td>Textured Si solar cell with lossless paper-based ARC</td>
<td>568.1 ± 0.3</td>
<td>28.91 ± 0.02</td>
<td>57.2 ± 1.0</td>
<td>9.39 ± 0.20</td>
</tr>
<tr>
<td>Enhancement</td>
<td>0.74 %</td>
<td>3.18 %</td>
<td>-0.19 %</td>
<td>3.76 %</td>
</tr>
</tbody>
</table>

2.2. Paper-based substrate for solar cells

The potential use of paper as a substrate for photovoltaic devices, driven by the desire to integrate solar cells on materials that need to be foldable (e.g., tents, clothes, etc.), ductile, lightweight, inexpensive, or environmentally friendly, has been investigated.\textsuperscript{[36, 57-65]} Organic photodiodes fabricated on regular newspaper showed a diode-like behavior in current-voltage (\(I-V\)) measurements.\textsuperscript{[57]} Even though the performance was not up to the mark for photovoltaic applications (\(i.e.,\) short-circuit current density, \(J_{SC}\), of 0.22 mA/cm\(^2\) and open-circuit voltage, \(V_{OC}\), of 0.4 V), this research demonstrates the feasibility of flexible optoelectronic devices on a variety of paper substrates. A solar cell using poly (3-hexylthiophene) (P3HT):1-(3-methoxycarbonyl)-propyl-1-phenyl-(6,6)C\(_{61}\) (PCBM) on a paper substrate was also made.\textsuperscript{[58]} However, the performance (\(J_{SC}\) of 0.1 mA/cm\(^2\) and \(V_{OC}\) of 0.39 V) needs improvement to make meaningful commercial products using this approach. Recently, a solar cell using cellulose nanocrystals (CNCs) as a substrate was introduced.\textsuperscript{[59]} CNCs are composed of fibers with width of 3 nm to 10 nm and length of 50 nm to 500 nm. The solar cell with the photo-
active layer of poly[(4,8-bis-(2-ethylhexyloxy)-benzo[1,2-b:4,5-b’]dithiophene)-2,6-diyl-alt-
(4-(2-ethylhexanoyl)-thieno[3,4-b]thiophene)-2,6-diyl]:PCBM (PBDTTT-C:PCBM) contacted by silver (Ag) electrodes demonstrates superior performance ($J_{SC}$ of 16.1 mA/cm$^2$, $V_{OC}$ of 0.68 V, fill factor, FF, of 54 %, and power conversion efficiency, PCE, of 2.7 %) in comparison to previous studies. However, these materials are still unable to match the performance of cells based on indium tin oxide (ITO) and glass substrates, as the transmittance is lower. Improved devices with higher PCE could be made by adjusting the properties of the CNC substrate (such as composition, orientation, interfaces, pH, etc.), by replacing the Ag contact with other materials having higher transmittance (such as metal-oxide, conducting polymer, etc.), by designing a novel cell structure, or by selecting more desirable active materials for solar cells.

3. Electronic circuits

Analog and digital electronic circuitry are two of the most important inventions in the past century, and it is hard to imagine the world without electronics. The electronic industry has been driven by the scaling, increased integration, and lower power consumption toward making highly efficient and low-cost devices. However, as the demand for flexible, lightweight, transparent electronic devices keeps growing, fabricating transistors on flexible and soft substrate has increasingly attracted researchers. For flexible electronic applications, paper has been extensively explored either as a substrate,[41, 66-93] or as a transistor component such as gate dielectric of field-effect transistors,[41, 94-97] tunnel layer for memory transistors,[98, 99], or as a dual-purpose layer (e.g., simultaneously act as both a substrate and a gate dielectric of field-effect transistors).[41, 92, 93] An insulator containing eco-friendly electrolytes derived from cellulose is another example of paper usage in transistors.[100]

3.1. Paper as a substrate for flexible devices
The use of paper as a substrate for making transistors for flexible and portable electronics is rather appealing. Organic FETs fabricated on NFC-based nanopaper have been recently demonstrated. The nanopaper with an average fiber diameter of ≈ 10 nm has a high optical transmittance and low surface roughness. A highly conductive single-walled carbon nanotube (SWCNT) film is deposited on the nanopaper as a gate electrode, followed by a coating of poly(methyl methacrylate) (PMMA) film as a dielectric layer. N-type semiconductor (F15-NTCDI (F15 = bis(pentadecafluorooctyl), NTCDI = naphthalenetetracarboxylic diimide) is the active FET layer on top of PMMA. A schematic in Fig. 4a shows the device structure, and a picture in Fig. 4b shows the bent device confirming its flexibility. It is important to note that the SWCNT film instead of a more common transparent conductive oxide (TCO) is used to make flexible devices. Both the nanopaper substrate and the fabricated FET remain transparent in the visible spectrum range (Fig. 4c). The electrical characteristics confirm good transistor behavior (Fig. 4d and e): from the measured I-V curve, effective carrier mobility and on/off modulation ratio are determined (4.3 × 10^{-3} cm^2/V·s and 200, respectively, in air at room temperature), and they are comparable to the majority of other n-type organic FETs. To assess the device’s long-term stability and its potential in wearable applications, bending tests were performed (Fig. 4f). While ≈ 10 % reduction of carrier mobility is observed, good device transfer characteristics are mainly preserved during the test.

Recently, a flexible low-voltage indium zinc oxide (IZO)-based homojunction thin-film transistor (TFT) using beeswax as a gate dielectric on a paper substrate has been introduced. Natural beeswax can be processed at a low temperature, and this layer possesses a high electric double layer (EDL) capacitance of 5 μF/cm^2 measured at 1 Hz. Due to its high EDL capacitance, the fabricated TFT can operate at a relatively low voltage of < 2.0 V. A high field-effect mobility of 14.6 cm^2/V·s, drain-source current on/off modulation ratio of 7.6 × 10^4, and a small subthreshold gate voltage swing of 86 mV per decade are
reported. Even without surface passivation, this TFT on a paper substrate shows excellent stability after 3,000 cycles of bending and 30 days of aging, which is confirmed by the minimal change of transfer characteristics.

The recently introduced oxide TFT on nanopaper has demonstrated appealing electrical properties. In general, these type of transistors require thermal annealing processes. However, there is a tradeoff between device performance and thermal treatment, which is a big challenge in fabricating oxide TFTs. To address this issue, a dual-layer channel comprised of aluminum oxide and indium gallium zinc oxide (Al₂O₃/IGZO) is employed to fabricate high-performance nanopaper TFT with conventional physical vapor deposition (PVD), which does not require conventional thermal annealing treatments. This novel nanopaper-based TFT demonstrates good electrical properties: a saturation mobility of 15.8 cm²/V·s, and an on/off modulation ratio of 4.4 × 10⁵, a threshold voltage of -0.42 V, and a subthreshold gate voltage swing of 0.66 V per decade.

3.2. Paper-based gate dielectric

Cellulose paper can also be used as a gate dielectric layer for flexible FETs. The schematic of the device is shown in Fig. 5b. In this case, two different cellulose fiber-based papers with different porous cavities were explored. These paper-based dielectrics showed superior performance (i.e., high field-effect mobility of > 30 cm²/V·s, drain-source current on/off modulation ratio of 10⁴, exhibiting a near-zero threshold voltage, a subthreshold gate voltage swing of about 0.8 V per decade (Fig. 5c)) as compared to traditional amorphous Si TFTs or oxide-based TFTs on glass or on Si substrates that require high-temperature (i.e., 200 °C to 300 °C) annealing. Furthermore, the performance remained unchanged for two months after device fabrication illustrating the environmental stability of the device. The possibility of scalability, low-cost manufacturing processes, and low power consumption due
to the low operating bias of the device makes it a strong candidate to construct future disposable electronics, such as paper displays, smart labels, smart packaging, etc.

Paper as a gate dielectric layer was also used in complementary metal oxide semiconductor (CMOS) circuits.\cite{92,93} In these studies, a fiber-based paper was used both as a gate dielectric layer and a substrate for n- and p-type TFTs. The dual use eliminates an additional transfer process. A CMOS inverter with paper-based n- and p-type TFTs was demonstrated showing a potential to implement paper-based analog and digital mixed-signal integrated circuits. The fabricated paper-based CMOS inverter is shown in Fig. 5d. Its transfer characteristic with the corresponding gain confirms its usability as a building block for complex paper-based circuits (Fig. 5e). Using more compact papers has led to a reduced leakage current that is comparable to devices based on polymer or glass substrates.\cite{93}

However inexpensive and comfortable a paper-based device may be, it will have no value if a reliable operation is not guaranteed. Even though some studies confirm the potential stability of paper-based devices, studies on the reliability remain important. Particularly for high performance applications such as in memory devices (e.g., as a tunneling barrier), a detailed study of environmental factors that may affect reliable operation, including humidity, moisture, temperature, UV radiation, impurities, etc., should be conducted.

4. **Transparent conductive electrodes**

Transparent conductive electrodes (TCEs) are necessary components of flexible optoelectronics including touch screens, interactive devices, and top electrodes for photovoltaics. Incorporating various conductive materials (e.g., carbon nanomaterials, metal nanomaterials, inorganic materials, or organic materials) into transparent paper enables the preparation of TCEs with excellent optical, mechanical, and electrical properties for a variety of optoelectronic applications.\cite{14,56,61,101-121} However, there exists a tradeoff between high conductivity and high optical transmittance of TCEs: adding large amounts of conductive
materials will result in higher conductivity, but it will decrease the optical transmittance. Thus, many efforts have been devoted to find an optimal balance for such applications. To make conductive paper, conductive materials are either deposited onto or embedded into cellulose paper. However, as traditional cellulose papers consist of micro-sized pulp fibers having large cavities of 10 µm to 50 µm, deposited conductive materials may spread unevenly forming bumpy boundaries and resulting in highly resistive, non-uniform conductive paper. To achieve lower resistance and better uniformity, nanopaper composed of 15 nm to 60 nm wide nanofibers appears to be a promising substrate. Deposited conductive materials remain on the nanopaper surface yielding a uniform conductive film with low resistance of \( \approx 0.4 \, \Omega/cm \).\(^{[14]}\)

4.1. **Transparent paper-based electrodes for LEDs and organic solar cells**

Highly transparent nanopaper was first explored as an environmentally friendly and renewable substrate to make TCEs for photovoltaic applications. A transparent nanopaper-based TCE for organic solar cells is one such example.\(^{[61]}\) It is made by depositing indium tin oxide, carbon nanotubes (CNTs), or silver nanowires (AgNWs) on the surface of the transparent nanopaper made of cellulose nanofibers. Another transparent nanopaper-based TCE embedded with AgNWs was also introduced showing outstanding optical and electrical properties.\(^{[107]}\) Here, an isotropic, thin, and conductive AgNW networks were laminated on the transparent nanopaper showing a high transparency (\( > 91 \% \)) and a low sheet resistance (\( \approx 13 \, \Omega \) per square).

To further improve the performance of solar cells fabricated on transparent and conductive nanopaper, it is important to prepare transparent nanopaper with a highly uniform surface. In this regard, recently, a new transparent nanopaper-based TCE utilizing cellulose nanofibers with a width of 15 nm is introduced that can be applied both to LEDs and solar cells.\(^{[118]}\) To make the transparent nanofiber paper conductive, silver nanowires with a diameter of 50 nm to 100 nm and a length of 5 µm to 10 µm are deposited atop the surface.
The total thickness of the silver nanowires-coated nanofiber paper was 15 µm to 20 µm. Despite the conducting film on top, a good optical transmission was observed: this is shown in a picture of Fig. 6a, where the conductive nanofiber paper is still transparent (right) and comparable to the transparent nanopaper without silver nanowires (middle). In both cases, incident light undergoes minimal scattering, which is distinct from a regular white paper (left) with numerous porous gaps resulting in significant light scattering. High optical transmission is quantified with the measurement shown in Fig. 6b: in the wavelength ranges of interest (λ = 300 nm to 800 nm), the measured transmissivity of the conductive nanofiber paper is ≈ 90 %. Next, the paper was evaluated as a transparent conductive electrode for LEDs and solar cells. Figure 6c shows that LEDs with conductive transparent nanopaper remain operational regardless of folding of the nanopaper, which illustrates the potential of this paper for flexible electronics applications (left: unfolded nanopaper, middle: folded nanopaper, right: recovered nanopaper after folding action). The nanopaper was also tested as an electrode of an organic solar cell having an active layer of P3HT/PCBM. The measured $I-V$ curve shows that the photo-generated carrier collection via the conductive transparent nanopaper is comparable to that of a cell with a more common indium tin oxide (ITO) electrode (Fig. 6d). Unlike ITO and glass-based conventional organic solar cells, the fabricated solar cell with conductive transparent nanopaper can be folded as needed, enabling the fabrication of portable, lightweight, and flexible solar cells (Fig. 6e).

4.2. Paper as a template for transparent conductive electrode

Another study demonstrated a transparent conductive electrode with good optoelectronic properties (i.e., sheet resistance of ≈ 2 Ω per square with transmittance of ≈ 90 %) and substantial mechanical flexibility that uses paper as a sacrificial template. The fabrication and the transfer process are shown in Fig. 7. Polymer nanofiber templates are made by electrospinning and coated with a material of choice (e.g., silicon, ITO, gold, silver,
copper, platinum, aluminum, chromium, nickel, or their alloys) using a standard thin-film deposition process. The coated fibers are then transferred onto substrates, such as a glass slide, PET, paper, textile, or a curved glass flask as shown in Fig. 7. Finally, the polymer nanofiber templates are dissolved in water or in organic solvents. The transmittance of metal nanotrough networks measured in a range of wavelengths from 300 nm to 2000 nm is nearly constant. Compared to ITO, the standard transparent electrode for solar cells, this performance is better in the near-infrared (NIR) range where the ITO becomes opaque. Furthermore, due to its wideband spectral performance, the metal nanotrough networks do not require additional chromatic correction components, which simplify the optical system design. As already mentioned in the prior section, to increase conductivity, a larger amount of conductive materials can be added to the paper template; however, this will decrease the optical transparency. Thus, finding an optimal operating condition for each application by balancing the two factors is an important design consideration for transparent and conductive paper-based electrodes.

5. LEDs for displays

Flexible displays have attracted considerable attention driven by huge industrial demand and enormous potential for further development, and paper has been explored as a component of LEDs for displays. Recently, organic light-emitting diodes (OLEDs) fabricated on a paper substrate have been demonstrated for flexible, lightweight, and thin displays.\textsuperscript{42,122-131} Minimizing surface roughness, improving organic solvent resistance, and increasing dimensional stability of paper are the most critical requirements for these applications. To reduce surface roughness while enhancing solvent resistance against organic solvents, the use of nanofibrils (\textit{i.e.}, having a diameter of $< 0.1$ μm) rather than microfibrils (\textit{i.e.}, having a diameter of $< 1$ μm) as paper constructing components appears to be helpful.\textsuperscript{24,172} Also, filling porous cavities within paper with polymers is effective in lessening the influence of
external factors (e.g., water, humidity, etc.) causing malfunctions and to improve paper shape stability.

5.1. Paper-based flexible OLEDs

In a recent study, OLEDs were fabricated on flexible cellulose nanocomposites with low coefficient of thermal expansion (CTE).\textsuperscript{[123]} Nine different matrix resins reinforced with cellulose nanofibers were evaluated. Transmittance of nanocomposites decreases by 6 \% to 9 \% compared to the transmittance of neat resins. However, the nanocomposites still transmit 82 \% to 85 \% of light in visible range, which is sufficient for a high-quality flexible OLED display as shown in Fig. 8a. In another study, OLEDs fabricated with similar approach, on a nanocomposite film composed of bacterial cellulose and polyurethane resin as a substrate, demonstrated good light-emitting behavior even when the device is bent (Fig. 8b).\textsuperscript{[124]} The nanocomposite, an environmentally friendly material showing more than 80 \% of optical transmittance in the visible spectral range and a high dimensional stability with low CTE of 18 \(\mu\text{m/m/K}\), is seen as a promising material for future flexible OLED displays.

5.2. OLEDs based on polymer-filled paper for better out-coupling efficiency

A recently demonstrated OLED fabricated on a plastic-paper hybrid substrate utilizes both high optical transmission (\(> 85 \%\)) and haze (\(> 90 \%\)) for higher luminescence and out-coupling efficiency.\textsuperscript{[42]} The structure of the OLED atop the plastic-paper hybrid substrate is shown in Fig. 8c. Paper is combined with plastic via templated polymer infiltration to reduce surface roughness of paper down to \(\approx 3\ \text{nm}\) that is essential for the multi-layer OLED deposition processes. The hybrid substrate has favorable optical properties (e.g., high transmission, high haze) that could not be achieved within a single-component material. The mesoporous air cavities within the cellulose paper are filled with polymer having a refractive index close to that of paper, thereby increasing transmittance. However, small angle light
scattering at numerous polymer/cellulose microfiber interfaces still exists, resulting in a high haze. Furthermore, this hybrid film, while remaining flexible, has a better shape stability compared to regular paper-based substrates as seen in Fig. 8d. The combination of favorable material, optical, and mechanical characteristics (i.e., uniform surface, high transmission, high haze, and good shape stability with high flexibility) results in improved power efficiency (> 35.1 % increase compared to OLED on plastic or glass substrate) and luminescence efficiency (> 6.6 % increase compared to OLED on plastic or glass substrate) when the light is emitted through the hybrid substrate. The improved out-coupling is attributed to the high haze with broad angle transmission of the hybrid substrate. The \( I-V \) characteristic and the light output efficiency of the fabricated OLED are shown in Fig. 8e. Electronic properties of OLEDs on multiple substrates including plastic, glass, and plastic-paper hybrid are compared and summarized in Table 3.

### Table 3. Summarized electronic properties of OLEDs on plastic, glass, and plastic-paper hybrid substrate.\(^{[42]}\)

<table>
<thead>
<tr>
<th>Device</th>
<th>Device 1 (cd/m(^2))</th>
<th>Device 100 (cd/m(^2))</th>
<th>Device 1000 (cd/m(^2))</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Voltage (V)</td>
<td>Current efficiency (cd/A)</td>
<td>Power efficiency (lm/W)</td>
</tr>
<tr>
<td>(1) OLED on plastic</td>
<td>2.8</td>
<td>63.7</td>
<td>62.7</td>
</tr>
<tr>
<td></td>
<td>3.8</td>
<td>61.4</td>
<td>52.3</td>
</tr>
<tr>
<td>(2) OLED on glass</td>
<td>2.8</td>
<td>66.6</td>
<td>69.7</td>
</tr>
<tr>
<td></td>
<td>3.8</td>
<td>65</td>
<td>56.8</td>
</tr>
<tr>
<td>(3) OLED on plastic-paper</td>
<td>3.2</td>
<td>90</td>
<td>74.3</td>
</tr>
<tr>
<td></td>
<td>4.8</td>
<td>91</td>
<td>59.8</td>
</tr>
<tr>
<td>Enhancement (3) vs. (1)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>41.3 %</td>
<td>18.5 %</td>
<td>48.85 %</td>
</tr>
<tr>
<td>Enhancement (3) vs. (2)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>35.1 %</td>
<td>6.6 %</td>
<td>40 %</td>
</tr>
</tbody>
</table>

6. Portable energy storage

Portable electronics have revolutionized our everyday life, and energy storage devices are among the enablers of these changes. To further capitalize on the enormous potential of this revolution, the issues of flexibility, weight, and the relatively high price of energy storage
must be tackled requiring intensive research and development efforts. Recently, paper has come into the spotlight as a promising component of energy storage devices due to its positive characteristics, such as environmental-friendliness, thinness, mechanical flexibility, lightweight, low-cost, and low thermal expansion coefficient. Various energy storage devices including batteries and supercapacitors might benefit using paper as electrodes,[108-114, 116, 132-147] separators,[101, 132, 134, 148, 149], reservoir,[150, 151] or substrate.[152] In energy storage applications, papers based on microfibrils or nanofibrillated cellulose (NFC) for electrolyte containment and for the control of the ion diffusion have significant advantages due to their mesoporous structure.

6.1. Supercapacitors with paper-based electrodes

Recently, a supercapacitor based on filter paper has been demonstrated.[144] The supercapacitor utilizes nickel (Ni)/manganese dioxide (MnO$_2$)–filter paper (FP) as a positive electrode, Ni/active carbon (AC)–FP as a negative electrode, and polyvinyl alcohol (PVA)–sodium sulfate (Na$_2$SO$_4$) as an electrolyte. Figure 9a illustrates the process utilized in making the flexible Ni/MnO$_2$–FP positive electrode. To construct the initial conductive networks, a homogeneous Ni layer is coated using a traditional electroless plating. To further improve the conductivity, the second layer of Ni is deposited by electro-deposition. Next, an electrochemically active material, MnO$_2$, is deposited by anodic electro-deposition as a final layer of the positive electrode for the supercapacitor. The completed solid-state flexible asymmetrical supercapacitor consists of positive and negative electrodes with PVA–Na$_2$SO$_4$ gel as an electrolyte. The schematic in Fig. 9b shows the supercapacitor connected to an LED. The fabricated device is mechanically stable and flexible, operating even when bent (Fig. 9c). The LED lights up powered by two supercapacitors (Fig. 9d). Cyclic voltammetry (CV) curves measured at a scan rate of 100 mV/s show roughly rectangular shape (Fig. 9e), which becomes larger as the potential windows increases from 1.0 V to 2.5 V. Based on the
measurement, both areal and volume capacitance over different potential windows (1.0 V to 2.5 V) are calculated (Fig. 9f). As the scan rate is reduced from 100 mV/s to 5 mV/s, an areal capacitance increases from 275 mF/cm$^2$ to 700 mF/cm$^2$. The capacitance is maintained above 85% of the initial value over 1,000 cycles at a current density of 20 mA/cm$^2$. The flexibility and thinness as well as the high performance of the supercapacitor show the possibility of developing wearable and portable applications with this technology.

A thin-film transparent supercapacitor using a hybrid paper based on cellulose nanofibers (CNFs) and reduced graphene oxide (RGO) nanosheets (i.e., CNF-[RGO]$_n$, $n = 10, 20$) has also been demonstrated.$^{[138]}$ In this case, the paper prepared by a layer-by-layer self-assembly technique is used as an electrode and charge collector. The capacitor has an areal capacitance of 1.73 mF/cm$^2$ at the current density of 0.012 mA/cm$^2$, while being partially optically transparent (56% at a wavelength of 550 nm). The good electrochemical characteristics including cyclic stability are attributed to the combination of anti-stacking of the distorted RGO nanosheets and internal nanoscale electrolyte reservoirs in the hybrid paper. Similar functionality can be achieved using transparent cellulose nanofibers as a substrate for supercapacitors.$^{[110]}$ As in the previous example, the multilayers are deposited using a layer-by-layer assembly of cellulose nanofiber paper, positively charged polyaniline (PANI) nanowires, and negatively charged graphene oxide nanosheets or poly(3,4-ethylenedioxythiophene: poly(styrene sulfonate)) (PEDOT:PSS) nanoparticles. Both structures, with reduced graphene oxide and PEDOT:PSS, show good electrochemical characteristics (i.e., 5.86 mF/cm$^2$ at the current density of 0.0043 mA/cm$^2$ for CNFs/[PANI-RGO]$_8$, and 4.22 mF/cm$^2$ at the current density of 0.0043 mA/cm$^2$ for CNFs/[PANI-PEDOT:PSS]$_8$) and have high cyclic stability. This performance is enabled by the synergetic effects of the electrical double layer capacitance of RGO and the pseudo-capacitive property of PANI and PEDOT:PSS. These results demonstrate a potential for thin-film supercapacitors based on cellulose nanofibers.
6.2. Li-ion batteries with paper-based anodes

A conductive nanopaper over-coated with Si has been explored as an anode of a lithium (Li)-ion battery.\textsuperscript{[139]} The anode is formed by carbon nanotubes (CNTs) interwoven with nanofibrillated cellulose (NFC). Because of a low percolation threshold of CNT networks, the nanocomposite conducts electrons even in a highly porous structure that can be filled with electrolyte. A thin layer of Si is then deposited by plasma-enhanced chemical vapor deposition (PECVD) on the conductive NFC:CNT nanocomposite, partly penetrating into porous cavities within the nanocomposite. The thin cross-section of this film enables faster Li-ion within the solid-state electrode resulting in higher power. This conductive nanopaper as an anode in a Li-ion battery shows a stable capacity (1200 mA·h/g for 100 cycles). As the paper is a flexible, lightweight, porous, and low-cost material with tunable properties, this result is encouraging for the development of large scale, low-cost energy storage devices.

7. Antennas

With multiple personal portable devices becoming a vital part of our everyday life, finding a way to make light, flexible, foldable, and cost-effective antennas for communication has become another major technological opportunity. In this regard, antennas fabricated on paper-based substrates\textsuperscript{[153-166]} offer boundless potential to satisfy the recent technology trends.

7.1. 3D paper antenna

A three-dimensional (3D) antenna patterned with a silver ink-contained ballpoint pen shows the potential of paper-based antennas in future communication devices.\textsuperscript{[159]} Radiating elements are drawn on paper by the ballpoint pen in meander line patterns with a width of 650 μm and spacing of 1 mm (Fig. 10a). To make an antenna, the patterned paper is transferred
onto a glass hemisphere having a radius of 12.7 mm, and then connected to a low-loss laminate substrate via copper feed lines. The reflected power measured with a network analyzer (Fig. 10b) is 4.9 dB at 1.85 GHz, and it can be improved if silver meander lines were cured at higher temperature (550 °C) to achieve better conductivity. However, the investigation of stability of cellulose at such high temperature also should be made. This work demonstrates the potential of paper-based antennas for communication devices.

7.2. **Foldable, tunable, and portable paper-based antenna**

Better performance was demonstrated by an antenna made of silver nanowires on a paper substrate consisting of cellulose nanofibers with a width of ≈ 30 nm. The foldability of this antenna provides a distinct advantage over rival designs, e.g. antennas fabricated on plastic substrates. The foldability also allows for tuning the frequency of return loss, which is not possible with conventional substrate-based antennas, thereby enabling transmission and reception of multiple frequencies. Figure 10c clearly shows the tuning of return losses depending on how much the paper-based antenna is folded. Furthermore, the sensitivity of paper-based antennas can be superior to that of plastic-based antennas. This seems to be counterintuitive because the high surface roughness of paper usually results in lower conductance and lower antenna sensitivity. However, paper compression can significantly reduce sheet roughness. By pressing the pulp paper with micro-sized cellulose fibers of 10 μm to 30 μm, the return loss was reduced from -7.4 dB to -26.7 dB. It can be further improved to -32.0 dB with a nanopaper substrate composed of 30 nm cellulose nanofibers as the surface roughness is further decreased by 15 to 80 times. This performance exceeds that of an antenna on a plastic substrate with a return loss of -30.1 dB. A similar antenna fabricated on a nanopaper composite with a high dielectric constant was demonstrated in targeting the same frequency range of Wi-Fi communication (i.e., IEEE 802.11n, operation on 2.4 GHz band) (Fig. 10d–g). This work shows the possibility of making high quality foldable antennas
for portable electronic devices. However, guaranteeing durability of paper under numerous folding actions remains the central issue for paper-based foldable antenna applications.

7.3. Paper-based antennas for RFID

Flexible antennas printed on a transparent nanopaper are also explored for radio frequency identification (RFID) applications. In this work, the cross-linking treatment is applied to increase the shape stability of the transparent nanopaper. During this process, the hydroxyl groups interact with the aldehydes resulting in the cross-linked structure. The cross-linked structure guarantees high shape stability of the transparent nanopaper during the solution-based (e.g., ink) printing process. RFID antenna patterns are ink-printed using a gravure press on the transparent nanopaper to demonstrate a low-cost fabrication. At a resonant frequency at 683.75 MHz, good RF performance is observed with insertion losses of -37.9 dB and -38.85 dB for the 100 and 200 low probability of intercept antennas, respectively.

8. Conclusion

Mobile, flexible, and individualized devices have been extensively developed in response to the growing demands of the modern life style. Selecting and applying materials with distinctive characteristics, such as tunable properties, easy modifications, cost-effectiveness, lightweight, flexibility, or environmental-friendliness, that can increase the utility, competitiveness of these devices and enable novel applications, has been a driver of research in the field. In this regard, paper has been spotlighted as a promising candidate material for future electronic and optoelectronic devices. In this review, various current electronic and optoelectronic applications using paper are introduced. Here are key summaries of characteristics of paper and relevant applications:
• For its foldability and flexibility, paper has been adapted as a substrate in many electronic and optoelectronic devices.
• Highly transparent paper made of either microscale cellulose fibers, cellulose nanofibers, or by the infiltration of pores within paper with a transparent material has been developed for diverse optoelectronic devices in which high transparency is required.
• By adjusting porous size, porosity, and/or fiber size of the paper, optical haze can be tuned while retaining high transparency, targeting a range of novel applications.
• Combining high transmission and high optical haze, cost-effective and environmentally friendly paper-based antireflection coatings can be made, which are particularly relevant for photovoltaics and OLEDs.
• A paper having high transmission and low optical haze is a promising substrate for diverse flexible display panel applications.
• A paper coated with conductive materials can be used as transparent conductive electrodes for various flexible devices including touch screens, interactive devices, and top electrodes for photovoltaics.
• Its intrinsic high resistance facilitates its application as an insulator in electronic devices including memory and field effect transistors.
• Due to their intrinsic mesoporous structure, paper is a good candidate material for electrolyte containment and for the control of the ion diffusion in energy storage devices.
• The foldability of paper enables an easy tuning of antenna’s operational bandwidths, which offers boundless potential to make lightweight, flexible, and cost-effective portable communication devices.

However, some obstacles remain for paper devices truly penetrating to everyday life. A lack of durability, both mechanical and environmental, is one of the major issues that needs to be addressed for the extensive use of paper in electronic and optoelectronic devices. As most devices should perform in realistic harsh indoor/outdoor environments, ways to make
robust paper while maintaining existing positive properties are the key research challenges. Stable material, electronic, optical, and optoelectronic properties must be maintained regardless of the frequency or number of folding events. Addressing other issues, such as preserving the shape stability, reducing the surface roughness, waterproofing, enhancing the barrier properties against oxygen or other reactants, etc., is also important for these applications. Further studies with paper to realize better performing optoelectronic and electronic devices should also be made. Particularly for photovoltaics, a multiple stack of paper-based antireflection coating can be considered for better optical coupling to solar cells. Either by changing pulp material or density, or by using different binding materials, the effective refractive index of paper can be modified. By constructing a multiple stack paper layer having gradual change of the refractive index, better performing antireflection coating can be achieved. For paper-based flexible electronics, careful study on the capability of electromagnetic shielding should be performed. As those devices cannot employ traditional solid material-based packages for electromagnetic shielding, confirming their resistant capability on electromagnetic interference would enable paper to be truly used in scaled down analog and digital mixed-signal integrated circuits. Cost-effectiveness is one of the main advantages of paper-based devices, however, significant research and development will be needed on the high-throughput roll-to-roll manufacturing process of paper-based electronic and optoelectronic devices by combining widely used manufacturing techniques, such as coating, printing, or lamination.

Acknowledgements

The authors acknowledge B. Hoskins and B. Natarajan for their fruitful comments. D. Ha acknowledges support under the Cooperative Research Agreement between the University of Maryland and the Center for Nanoscale Science and Technology at the National Institute of Standards and Technology, Award 70NANB14H209, through the University of Maryland. Z. Fang acknowledges the Young Scientists Fund of the National Natural Science Foundation of China (Grant
No. 31700508), the Natural Science Foundation of Guangdong Province, China (Grant No.2017A030310635), the State Key Laboratory of Pulp and Papermaking Engineering (2016PY01, 201709), and Open Funds of National engineering research center of near-net-shape forming for metallic materials (2016006).

Received: ((will be filled in by the editorial staff))
Revised: ((will be filled in by the editorial staff))
Published online: ((will be filled in by the editorial staff))

References


**Figure 1.** (a) Schematic showing the hierarchical structure of a tree. (b–c) Schematic illustrating structure of (b) a regular paper and (c) TEMPO-oxidized wood fibers and the corresponding chemical structures of the constituent cellulose. Reproduced with permission.\textsuperscript{[36]} Copyright 2013, American Chemical Society. (d) SEM images of a regular paper and TEMPO-oxidized transparent paper. The transparent paper shows fewer porous air cavities and thus higher packing density of cellulose fibers than the regular paper, resulting in high transmission.
Figure 2. (a–c) Paper transfer and removal process. (a) The transparent cellulose paper used as an antireflection coating. (b) Transfer of the transparent paper antireflection coating on a GaAs solar cell. PVAc is used as a binding material. (c) The cell with the transparent cellulose paper antireflection on top. (d) The paper can be removed as needed. (e) A SEM image showing cellulose fibers within the transparent paper antireflection coating. Reproduced with permission.[53] Copyright 2014, Wiley-VCH.
Figure 3. (a–d) Measured and calculated optical properties with and without a paper-based antireflection coating on a GaAs solar cell. (a) Measured reflectivity of a bare GaAs solar cell and (c) measured reflectivity of the same device with the transparent paper antireflection coating on top. (b) Calculated reflectivity for a bare GaAs substrate and (d) calculated reflectivity with the transparent paper antireflection coating layer on top of the GaAs substrate. All measurements and calculations are made as a function of incident angle and wavelength. Measurements are performed using an integrating sphere and monochromatic light. Calculations are made based on incoherent reflection model accounting for multiple
reflections within the layer. Calculations satisfactorily agree with the experimental data. (e–f) Measured electronic properties with and without the paper-based antireflection coating on a GaAs solar cell. (e) EQE is measured over the operational spectrum of a GaAs solar cell. Blue solid line indicates measured EQE of a bare GaAs solar cell. Two different EQE measurements with the paper-based ARC (red solid and dotted lines) confirm improved electronic characteristic induced by absorptivity enhancements. (f) Current-voltage characteristics with (red solid line) and without (blue solid line) the paper-based ARC are determined. Increased light absorption within the active material improves the short-circuit current density by ≈ 20 %, which in turn results in ≈ 24 % enhanced power conversion efficiency with a little enhancement in the fill factor. Reproduced with permission.[53] Copyright 2014, Wiley-VCH.
Figure 4. (a) Schematic showing the organic FET with a nanopaper substrate. Inset molecular structure shows F15-NTCDI semiconductor. (b) A picture of the real fabricated device when it is bent. (c) Measured optical transmittance of nanopaper used in this study and the fabricated device (≈ 89 % and ≈ 84 % at a wavelength of 550 nm, respectively). (d) Measured drain-source current-voltage characteristics ($I_{SD}$-$V_{SD}$) of the device for each applied gate voltage from 0 V to 80 V. (e) Transfer characteristic of the device when $V_{SD}$ = 10 V is measured. Inset shows how much gate leakage current is measured under gate voltage. (f) Transfer characteristics of the device when it is bent. Black solid line, red dashed line, and blue dashed line represent measured transfer characteristics when the device is not bent, bent in vertical to the direction of conduction channel, and bent in parallel to the direction of conduction channel, respectively. Reproduced with permission.[77] Copyright 2013, American Chemical Society.
Figure 5. (a) SEM images showing two different types of papers. Type A has more porous cavities while type B is highly packed. (b) Schematic of the field effect transistor using cellulose paper as a gate dielectric. (c) $I$-$V$ transfer characteristics measured at saturation region (drain voltage of 15 V) for devices with two different types of papers. Measurements are conducted at room temperature. Reproduced with permission. Copyright 2008, Institute of Electrical and Electronics Engineers. (d) An image showing paper-based CMOS inverter with p-type TFT (a gate width to length (W/L) ratio of 21.8) and n-type TFT (W/L ratio of...
11.8). (e) Device transfer characteristic of the paper-based CMOS inverter with the corresponding gain. Reproduced with permission.[93] Copyright 2013, Wiley-VCH.

Figure 6. (a) A picture showing high optical transmittance of the nanopaper with and without silver nanowires. Left, middle, and right arrow indicates a regular white paper, a nanopaper without silver nanowires, a nanopaper with silver nanowire coatings atop, respectively. (b) Measured optical transmittance of the nanopaper with (black dots) and without (black solid line) silver nanowire coatings. (c) LED operation with the conductive transparent nanopaper. Left, middle, and right pictures show the LED operation when the nanopaper is unfolded, folded, and recovered from folding action, respectively. (d) Measured electronic characteristic
of an organic solar cell with the conductive transparent nanopaper as an electrode (illumination: red solid, dark: red dots) and with ITO as an electrode (illumination: black solid, dark: black dots). (e) The fabricated organic solar cell with the conductive transparent nanopaper electrode is flexible, lightweight, and portable. It can supply electricity everywhere. Reproduced with permission. Copyright 2015, Creative Commons License.

Figure 7. (a) Schematic showing the fabrication process of nanotroughs. Polymer nanofiber web made by electrospinning is coated with selected materials. Once the fibers are coated, they are laminated on substrates, and then polymer-fiber template is removed by immersing it in water or in an organic solvent. (b) Left: A SEM image showing gold nanotrough networks.
Right: A SEM image showing a junction between two different nanotroughs. (c) Cross-sectional view of a SEM image of a gold nanotrough. (d) Prepared gold nanotroughs can be transferred onto diverse substrates. Left to right: Pictures showing gold nanotroughs on a glass slide, PET, paper, textile, and a curved glass flask, respectively. Reproduced with permission.\textsuperscript{[103]} Copyright 2013, Macmillan Publishers Limited.

\textbf{Figure 8.} (a) A picture showing an operation of an OLED based on a flexible and transparent cellulose nanocomposite with low coefficient of thermal expansion. Reproduced with permission.\textsuperscript{[123]} Copyright 2009, Elsevier Ltd. (b) A picture showing an OLED on bacterial cellulose nanocomposite. The fabricated OLED can emit light even when it is bent. Reproduced with permission.\textsuperscript{[124]} Copyright 2011, Elsevier B.V. (c–e) The OLED on plastic-paper hybrid substrate. (c) Schematic showing the layer information of the device. (d) The OLED atop plastic-paper hybrid substrate operates well when it is bent, confirming its shape stability and flexibility. (e) Current and light output efficiency are determined as a function of operation voltage. Reproduced with permission.\textsuperscript{[42]} Copyright 2016, The Royal Society of Chemistry.
Figure 9. (a) Schematic showing the fabrication process of a flexible Ni/MnO$_2$–FP positive electrode. (b) Schematic showing the device constructed with a Ni/MnO$_2$–FP positive and a Ni/AC-FP negative electrodes with PVA/Na$_2$SO$_4$ electrolyte. (c) A picture shows the device’s mechanical stability. (d) A picture shows an LED illumination by two supercapacitors in series connection. (e) Measured CV curves confirming the good electrochemical characteristic of the paper electrode-based supercapacitor. Measurements are confirmed at a scan rate of 100 mV/s when potential windows are varying from 1.0 V to 2.5 V. (f) Areal and volume
capacitance are determined from the measured CV curves under different potential windows. Reproduced with permission. Copyright 2015, American Chemical Society.

Figure 10. (a–b) A 3D antenna based on paper substrate. (a) A picture showing the fabricated paper-based antenna patterned with a silver ink-contained ballpoint pen. (b) Measured reflected power confirms communication capability of the paper-based antenna. Reproduced
with permission.\textsuperscript{[159]} Copyright 2011, Wiley-VCH. (c) Return losses of paper-based foldable antennas can be tuned depending on how much the paper is folded. Solid line shows the return loss of an unfolded antenna and dotted line shows that of a folded antenna. Reproduced with permission.\textsuperscript{[160]} Copyright 2013, The Royal Society of Chemistry. (d–g) A paper-based antenna for Wi-Fi communication. (d) Pictures showing a 30 mm long dipole antenna atop 50 µm thick nanopaper (left) and a 17 mm long dipole antenna atop 50 µm thick silver nanowire/nanopaper composite (right). (e) Return losses of the 30 mm long dipole antenna (solid line) and 17 mm long antenna on the silver nanowire/nanopaper composite (dashed line). (f) Pictures taken during bending of the nanopaper antenna and when it is restored. (g) The return loss of the nanopaper antenna before (solid line) and after (dashed line) 1,000 cycles of bending test. Reproduced with permission.\textsuperscript{[162]} Copyright 2014, Wiley-VCH.

**Author Biographies**

Dongheon Ha received an Honors B.S. degree in Computer Science and Electrical Engineering (double degree) from Handong University, Korea, a M.S. degree in Electrical Engineering from Korea Advanced Institute of Science and Technology (KAIST), Korea, and a Ph.D. degree in Electrical Engineering from the University of Maryland. His doctoral research focused on fundamental interaction between light and materials including dielectric, semiconductor, metal, etc. He worked as a researcher at SK Hynix Semiconductor to develop novel semiconductor devices and circuits. Dongheon is currently working as a Postdoctoral Researcher at the Center for Nanoscale Science and Technology (CNST), National Institute of Standards and Technology (NIST). He researches on nanoscale optical and electrical properties of semiconductor devices and also develops novel nanoscale measurement techniques.

Zhiqiang Fang received his M.S. and Ph.D. degrees in Pulp and Papermaking Engineering from the South China University of Technology (SCUT). From 2012 to 2013, Zhiqiang worked as a Visiting Scholar at the University of Maryland developing transparent paper for flexible electronics with Prof. Liangbing Hu. He also worked as a Postdoctoral Fellow in Prof. Honglong Ning's group in the School of Materials Science and Engineering at SCUT. Zhiqiang is currently working as an Assistant Professor at SCUT. His research interests include design and fabrication of multi-scale cellulose-based materials for green flexible electronics.
Nikolai B. Zhitenev received an Honors M.S. degree in Physics from the Moscow Institute of Physics and Technology, Russia, and a Ph.D. degree in Condensed Matter Physics from the Institute of Solid State Physics, Russia. Nikolai was a staff member at Bell Laboratories, Lucent Technologies, where his research focused on electronic transport in different physical systems, ranging from two-dimensional electron gasses in Si, Ge and GaAs, to semiconductor and metal quantum dots, to nanoscale molecular and polymer devices. As a Group Leader at the CNST, NIST, Nikolai leads multiple projects related to the nanoscale measurements of electronic and optical properties of novel materials and devices.