Highly Transparent and Flexible Nanopaper Transistors

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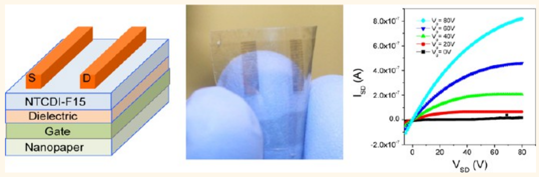
ABSTRACT Renewable and clean "green" electronics based on paper substrates is an emerging field with intensifying research and commercial interests, as the technology combines the unique properties of flexibility, cost efficiency, recyclability, and renewability with the lightweight nature of paper. Because of its excellent optical transmittance and low surface roughness, nanopaper can host many types of electronics that are not possible on regular paper. However, there can be tremendous challenges with integrating devices on nanopaper due to its shape stability during processing. Here we demonstrate for the first time that flexible organic field-effect transistors (OFETs) with high transparency can be fabricated on tailored nanopapers. Useful electrical characteristics and an excellent mechanical flexibility were observed. It is believed that the large binding energy between polymer dielectric and cellulose nanopaper, and the effective stress release from the fibrous substrate promote these beneficial properties. Only a 10% decrease in mobility was observed when the nanopaper transistors were bent and folded. The nanopaper transistor also showed excellent optical transmittance up to 83.5%. The device configuration can transform many semiconductor materials for use in flexible green electronics.



KEYWORDS: green electronics · nanopaper · transparent electronics · flexible

Cost-efficient and environmentally friendly macroelectronics, optimized with green materials and large-scale roll-to-roll processes, are attracting intensive research and commercial interests because they enable a range of disposable devices for consumer electronics.[1−7] Solution-based printing is an ideal process due to its fabrication speed: up to 15000 sheets per hour for offset printing. This process also boasts ideal scalability and benefits from techniques and equipment supporting a well-established paper industry. Conductors, thin film transistors, organic light-emitting diodes, and solar cells are already demonstrated with various printing methods. Previous efforts on printable electronics have generally focused on the development of new inks, fabrication processes, devices with new nanomaterials, and new device structures. However, printed devices are necessarily built upon substrates whose properties play an underappreciated role in device design, processing, and performance. Despite their important role, the development of new substrates is still lacking.

The interactions between the substrates, the metal contacts, and the active semiconductors greatly impacts the mechanical performance and sometimes the optical and electrical properties of devices.[8,9] Porosity, surface energy, defects, and flexibility are among the important parameters of substrates that play a key role in the printing process for printable devices. For example, it is much easier to print on cellulose paper than most other substrate materials due to its porous nature and excellent hygroscopy. Plastic, however, relies purely upon ambient evaporation for the solvent removal and immobilization of the electronic material. Glass substrates are widely used for electronics and displays, but sheet-by-sheet processing is used instead of roll-to-roll processing. Intensive research has already been conducted on the development of emerging substrates, especially plastic substrates with better humidity and oxygen barrier properties, better thermal stability, and a higher glass transition temperature. Corning Inc. recently developed flexible glass that has generated much interest. The flexible glass with 100 μm thickness

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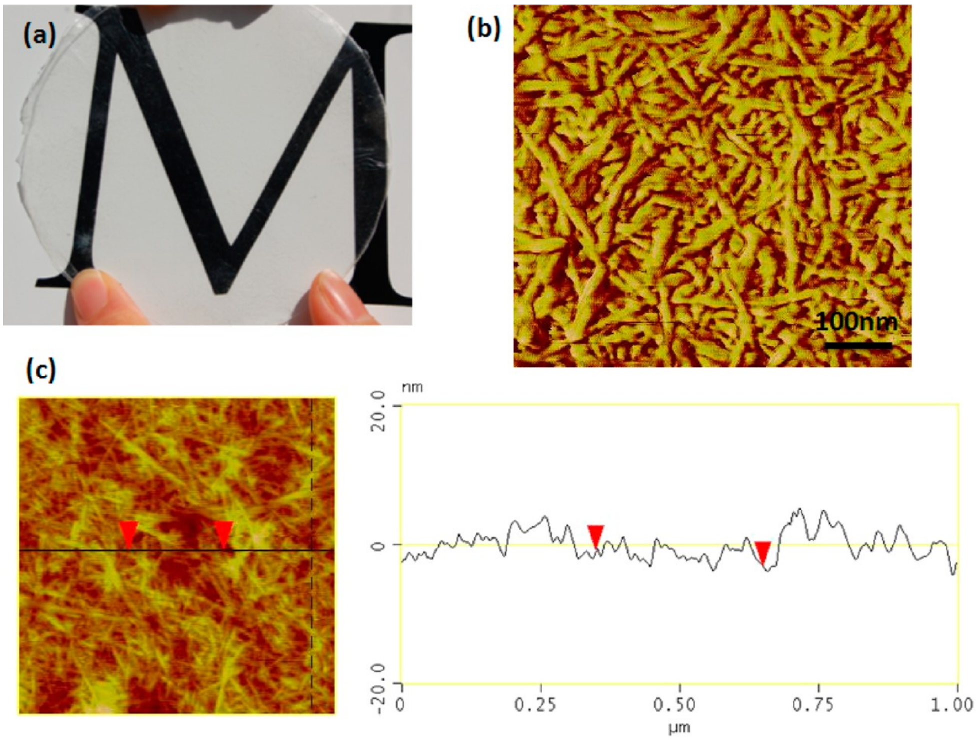

Figure 1. (a) Picture of a transparent nanopaper on top of "M". (b) AFM phase profile of porous nanopaper in (a) made from nanofibrillated cellulose with diameter ~10 nm. (c) AFM line scan of nanopaper in (a) with RMS 1 nm.

can be bent down to a 40 mm arc radius. The flexible glass is also highly transparent and can tolerate much higher processing temperature (600 °C) than plastic. This flexibility needs to be further improved to enable additional applications of the glass by reducing the minimum bending radius.

Nanopaper based on nanofibrillated cellulose (NFC) is an environmentally friendly and renewable material. Nanopaper is made of the same material as traditional paper but consists of fibers with a much smaller diameter than that of traditional paper. Reducing the diameter of the paper fibers decreases the optical scattering; therefore the nanopaper has excellent optical transparency. In addition, nanopaper has much better thermal stability compared to plastic substrates, with a coefficient of thermal expansion (CTE) of 12–28.5 ppm K^{-1}. The typical CTE value for plastic substrates is 20–100 ppm K^{-1}.[10] Nanopaper can also tolerate a much higher processing temperature than plastic.[10] The high transparency and flexibility of nanopaper allows to replace for plastic substrates in a wide range of applications. The field-effect transistor (FET) is an important component for many electronic devices, but to our knowledge there is no prior report of FETs successfully fabricated on nanopaper substrates. Here we are the first to demonstrate organic FETs designed and fabricated on this emerging substrate with excellent optical and mechanical properties. The transistor's performance, including its mobility and on/off ratio, may be further improved by optimizing the device structure and the fabrication process on the nanopaper substrate. When our nanopaper transistors were bent, only a slight decrease in mobility was observed, demonstrating the good flexibility for ultimate device applications.

RESULTS AND DISCUSSION

Nanopaper with high transmittance, low surface roughness, and excellent flexibility was prepared by vacuum filtration of an NFC solution. Bleached Kraft softwood fibers were pretreated with a NaBr/NaClO/TEMPO ((2,2,6,6-tetramethylpiperidin-1-yl)oxidanyl) system. TEMPO is a highly selective oxidant for the hydroxymethyl group at the glucose C6 position within the cellulose chain.[11,12] The swelled fibers were disintegrated to nanofiber with a diameter of 10–100 nm using a homogenizer (microfluidizer processor M-110EH). The obtained NFC suspension was filtered by a nitrocellulose ester filter membrane (Millipore DAWP29325) with 0.65 μm pore size. After filtration, the cellulose gel "cake" was placed between two smooth substrates and a polyamide woven fabric. Then it was hot pressed under high pressure to obtain low surface roughness and small pore size. Figure 1a shows the image of a transparent nanopaper made from nanocellulose fibers with an average fiber diameter of ~10 nm. When the nanofibrillated cellulose was dried from the water suspension, strong interfibrillar interactions were formed, including chemical hydrogen bonding from the large content of hydroxide groups and mechanical winding force between the different fibers. The mechanical properties of nanopaper are preserved even when nanopapers are made to be conductive by integrating polymers into the structure. Compared to traditional papers, nanopaper not only is mechanically stronger but also possesses higher transmittance, higher density, lower CTE, higher surface smoothness, more tunable structure, and better electrochemical properties in conductive composite.[36] All of these special properties contribute to making nanopaper a promising substrate for transparent and flexible electronics. Figure 1b and 1c show the atomic force microscopy (AFM) phase and height image of nanopaper, respectively. From the line scan of the surface height image in Figure 1c, we see the nanopaper has a low surface roughness, with maximum roughness depth R_{max} of 5 nm and root mean square roughness (RMS) of 1 nm. This is important for preventing a short leakage between the bottom gate electrodes and the semiconductor layer in FETs fabricated on nanopaper.

TABLE 1. Comparison of Nanopaper, Traditional Paper, and Plastic[10,20,21]

characteristics	nanopaper	traditional paper	plastic
surface roughness (nm)	5	5000−10000	5
porosity (%)	20−40	50	0
pore size (nm)	10−50	3000	0
optical transparency at 550 nm (%)	90	20	90
max loading stress (MPa)	200−400	6	50
coefficient of thermal expansion (CTE) (ppm K^{-1})	12−28.5	28−40	20−100
printability	good	excellent	poor
Young modulus (GPa)	7.4−14	0.5	2−2.7
bending radius (mm)	1	1	5
renewable	high	high	low

Recently, cellulose papers have been explored to replace plastic substrates as a lightweight substrate for low-cost, versatile, and roll-to-roll printed electronics.[13] Various types of devices such as transistors, radio frequency identification (RFID), light-emitting diodes, solar cells, paper based batteries, supercapacitors, and porous magnetic aerogels have been demonstrated on low-cost, recyclable regular paper.[14−18,35,37] Nanopaper has many advantages over both regular paper and plastic substrates, as shown in Table 1. Most notably, light goes through the fibers without scattering when the fiber size and spacing are both much smaller than the wavelength of light, making the material highly transparent. The light scattering scales with the fiber diameter proportional to $\sim D^3$, where D is the diameter of the fiber.[19] The nanopaper made from 10 nm NFC and densely packed with low surface roughness possesses significantly reduced surface and bulk scattering.

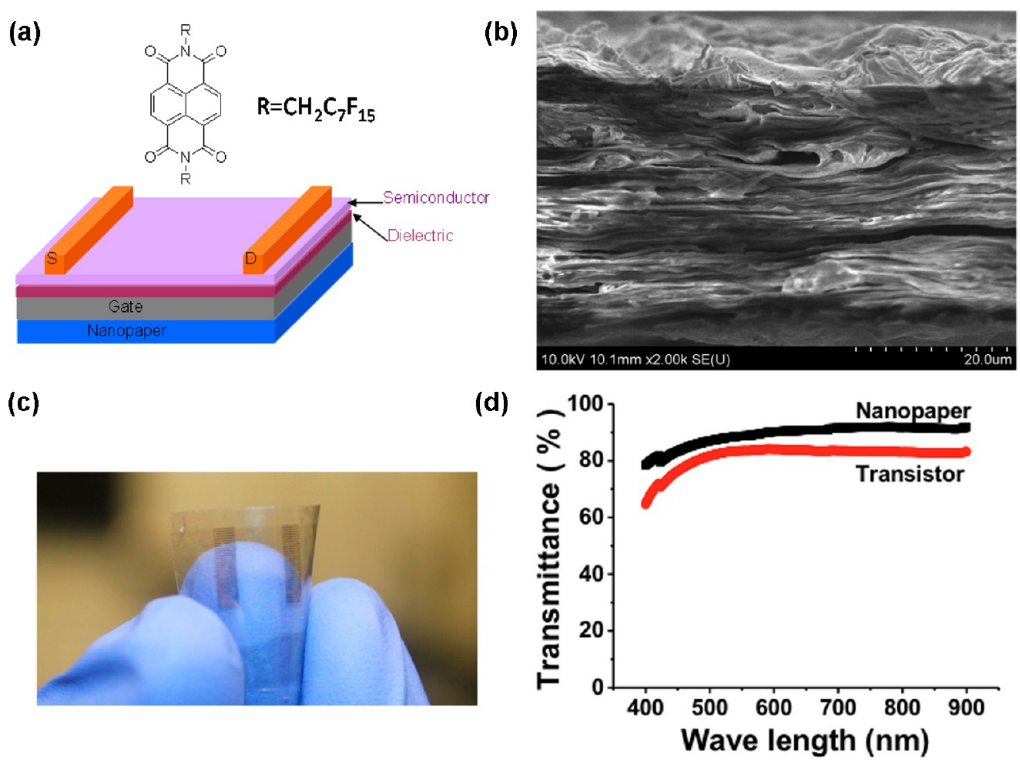

Figure 2. (a) Molecular structure of NTCDI-F15 semiconductor and a schematic drawing of the nanopaper OFET with a top-contact geometry. (b) SEM cross-section of a nanopaper transistor with a layered structure. Scale bar is 10 μm. (c) A picture of a fabricated transparent and flexible nanopaper transistor. (d) Optical transmittance of a nanopaper and nanopaper transistors with transmittance of 89% and 84% at 550 nm, respectively.

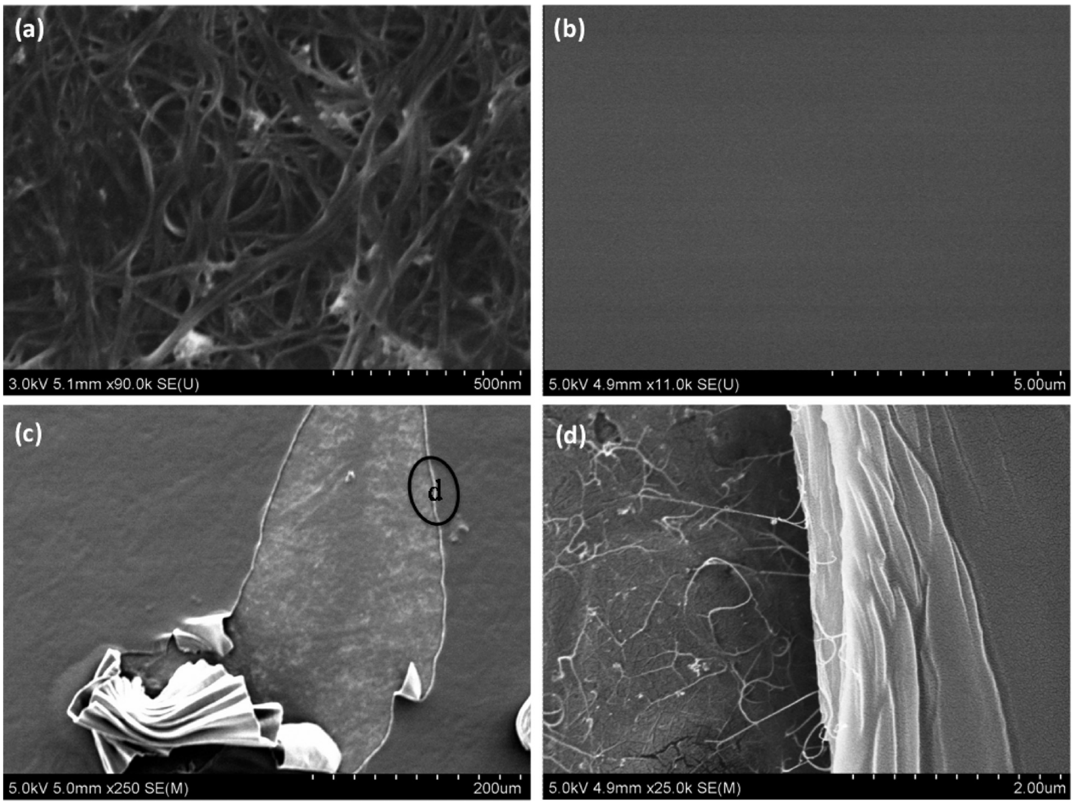

Figure 3. (a) SEM image of CNT film serving as the gate electrode on nanopaper. (b) SEM image of PMMA coated nanopaper and CNT film. (c) SEM image of a peeled spot at PMMA layer, and (d) zoomed in SEM image of the circled area in (c).

This makes transparent electronics on paper a realized concept. Other important advantages of nanopaper for electronic devices include the extremely low surface roughness, suitable pore size, and porosity. These properties make it possible to deposit flexible conductors such as single-walled carbon nanotube (SWCNT) films by liquid processing on nanopaper. Meanwhile, the high Young modulus (from 7.4 to 14 GPa)[10,20,21] of nanopaper provides excellent mechanical stiffness as an electronic substrate. Nanopaper is more environmentally friendly than plastic substrates because it is made from 100% wood cellulose rather than a nonrenewable petroleum dependent source. Therefore, unlike plastic electronics, disposable nanopaper electronics will not contribute to *white pollution*, the name given in press to the occurrence of long-lived plastic bags discarded into the environment. At the same time, nanopaper has good ink adsorption property due to its 3D fiber structure and it is more suitable for large-scale roll-to-roll printing than many other substrates.

The unique optical and mechanical properties of nanopaper offer it great potential in various applications, especially in cost-efficient, transparent, and flexible electronics. The transistor is a fundamental building block of many electronic devices. To explore the potential of nanopaper in electronic applications, we have designed and demonstrated the fabrication of flexible and highly transparent OFETs on nanopaper. To keep the high transparency of the device, the semiconductor materials also need to be transparent. However, most organic semiconductors such as pentacene and polythiophene are strong light absorbers and are therefore unsuitable for this purpose. Here we used a n-type organic semiconductor NTCDI-F15, a naphthalenetetracarboxylic diimide derivative which is highly transparent in visible light and has relatively good stability in air.[22] Figure 2a shows the schematic diagram of the flexible and transparent field-effect transistor fabricated on nanopaper and the molecular structure of the transparent organic semiconductor NTCDI-F15. SWCNTs (Carbon Solutions Inc.) were deposited on the nanopaper substrate by a scalable Meyer Rod coating method, followed by drying in a vacuum oven.[23−26] The highly conductive CNT film serves as the transparent gate electrode of the transistor. A CNT film is used here instead of a transparent conductive oxide (TCO) film because TCO is brittle and can crack during the fabrication process, as shown in the Supporting Information (Figure S1). In addition, CNT film can be deposited by various low-cost methods such as rod coating and simple drawing methods (Supporting Information Figure S1), while the deposition of high quality TCO film usually requires expensive methods such as vacuum deposition or high temperature annealing. A poly(methyl methacrylate) (PMMA)

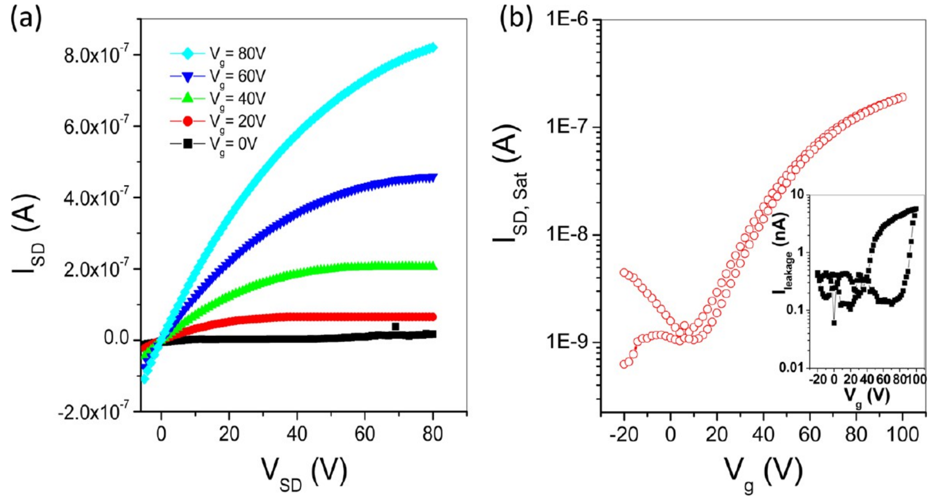

Figure 4. (a) $I_{SD}-V_{SD}$ characteristics of a nanopaper transistor. (b) Transfer characteristics of a nanopaper transistor. V_{SD} = 10 V. Inset shows the gate leakage current vs gate voltage.

dielectric layer was deposited on the nanopaper surface by spin-coating, followed by vacuum deposition of a NTCDI-F15 semiconductor film. Hitachi SU-70 scanning electron microscopy (SEM) with a JEOL JXA 840A system (JEOL Ltd.) was used for device morphology characterization. Figure 2b shows the SEM image of the device's cross section, which illustrates the nanopaper's layered structure. The nanopaper tends to self-assemble into a layered structure, which can effectively release bending strain, thus providing devices with good flexibility.[34] As shown in Figure 2c, the finished devices are highly flexible and transparent. To further confirm the optical transmittance of the fabricated devices, optical transmission spectra of both the nanopapers and the finished devices were measured by a Lambda 35 spectrophotometer (PerkinElmer, USA). Figure 2d compares the optical transmittance of the nanopaper and the finished transistor devices, both displaying very good transparency. The transmittance of the transistor at 550 nm is slightly reduced from the initial nanopaper's 89–84%, which is due to the additional layers of SWCNT, PMMA, and NTCDI-F15 film.

The microstructures and morphologies of each layer of active materials in a transistor impact its electrical and mechanical properties and hence affect the device's performance. SWCNT films are used as the gate electrode to provide high flexibility and optical transparency; however, electrical shorting due to the protruding CNTs is widely reported in devices such as transistors, organic solar cells, and light-emitting diodes.[27−29] Electrical breakingdown, self-healing, and multilayer dielectric blocking layers are various methods have been applied to solve this problem. Because of the strong interactions between CNTs and nanopaper fibers with rich surface functional groups, such as van der Waals forces and hydrogen bonding, CNTs tend to conformally stick to the fiber surface, which diminishes the number of protruding tubes.[30] SEM was performed on the SWCNT coated nanopaper with and without PMMA coating. As shown in Figure 3a, a uniform layer of SWCNT was coated on the nanopaper and served as the gate electrode. Protruding tubes are observed much less often than in a plastic substrate.[29] The smooth morphology shown in Figure 3b suggests that the SWCNTs were well covered by the PMMA dielectric layer after the spin-coating of PMMA. The thickness of the PMMA dielectric layer is controlled at ~1 μm. No pinholes were observed due to the smooth surface of the nanopaper. Note that the surface roughness of the nanopaper is ~5 nm, 3 orders of magnitude lower than regular paper. To further illustrate the morphology of the PMMA–CNT–nanopaper layered structure, the PMMA film was peeled off from the nanopaper in a selected area (see Figure 3c,d). As shown in Figure 3d, the SWCNTs can be seen in the peeled-off area, while Figure 3b demonstrates the clean unpeeled area surface.

The performance of the proof-of-concept transparent and flexible nanopaper transistors was evaluated by electrical characterization. The fabricated nanopaper OFETs exhibit good n-type transistor characteristics, as shown in Figure 4. The devices show obvious linear regime and saturation regime. Effective carrier mobility and I_{on}/I_{off} ratio was calculated from the $I_{SD}-V_{SD}$ (Figure 4a) curves and the transfer characteristic curves (Figure 4b), respectively. The saturation drain currents at different gate voltage were extracted from Figure 4a and then plotted into a $I_{SD}^{1/2}-V_g$ curve to calculate the mobility. When all measurements were performed in air, devices exhibited effective carrier mobility around 4.3×10^{-3} cm^2/(V s) and I_{on}/I_{off} ratio up to 200. The OFET mobility is comparable to that of many other n-type OFETs, with semiconductors deposited on substrates held at room temperature and measured in air. Heating the substrates during semiconductor deposition is expected to significantly enhance the mobility of the devices. The I_{on}/I_{off} ratio of

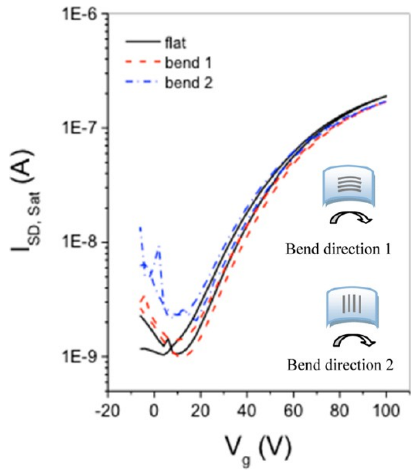

Figure 5. Transfer characteristics of nanopaper transistors before and during bending. The solid black curve shows the transfer curve of a nanopaper transistor measured on a flat surface before bending. The red and blue dashed curves were obtained from the bent device with bending direction vertical and parallel to the conduction channel direction. The bending radius is 3.5 mm.

the device is limited by a relatively high off-current I_{off}, which is partially due to the gate leakage current. Protruding CNTs may still be present on the nanopaper's surface, although the number density of SWCNTs penetrating the PMMA dielectric layer is significantly reduced due to the strong interactions between the nanopaper and the CNTs. Further optimization on the dielectric layer is needed to improve the I_{on}/I_{off} ratio.

Electronic devices with good mechanical flexibility enable a wide range of interesting applications such as wearable systems for personal health monitoring and smart gloves with integrated sensors.[1,2,31−33] To demonstrate the flexibility of our nanopaper transistors, the devices were wrapped around a vial and then electrical characterization was carried out in air during bending. Figure 5 compares the transfer characteristics of the nanopaper device before and during bending (bending radius = 3.5 mm). The solid black curve shows the transfer characteristics of a nanopaper transistor measured on a flat surface before bending. The red and blue dashed curves were obtained from the bent device with a bending direction vertical and parallel to the conduction channel direction, respectively. When the device was bent in the direction parallel to the conduction channel, as shown in the inset of Figure 5, a 10.2% decrease in mobility was observed. A 9.8% mobility reduction was observed when the device was bent in the direction vertical to the conduction channel. Note that the transistor is measured while it was bent, not once it recovered back to its original flat shape. Although the device structure was not designed nor optimized to reduce strain on the device's semiconductor layers and electrodes during the bending test, the device's transfer characteristics only changed a little when the device was bent. This result shows the good flexibility of nanopaper transistors, which is essential for flexible electronics. As shown in Figure 2b, the nanopaper tends to self-assemble into a layer-by-layer structure, which can effectively release the strain when the nanopaper is bent.[34] The conformal coating of the PMMA dielectric layer on the very smooth surface of the nanopaper may also contribute to the flexibility of the device because a smooth and uniform dielectric layer can effectively reduce the possibility of dielectric breakdown during bending. To further improve the flexibility of nanopaper transistors, many strategies can be applied. A example is using flexible nanowires or nanotube films as the source-drain electrode materials, and optimizing the device's structure so that the active semiconductor layer and electrode−semiconductor interface can be located close to the "strain neutral" position.[9]

CONCLUSIONS

In summary, nanopaper with optimal transmittance and surface smoothness was prepared based on nanostructured cellulose fibers. Nanopaper shows much lower surface roughness and much higher transparency than traditional paper. Highly transparent and flexible OFETs were successfully fabricated on the properly designed nanopaper. The nanopaper OFETs exhibit good transistor electrical characteristics. To demonstrate the flexibility of nanopaper OFETs, devices were measured before and during bending. Only a 10.2% and a 9.8% decrease in mobility were observed when the device was bent in the direction parallel to the conduction channel direction and vertical to the conduction channel direction, respectively. These excellent optical, mechanical, and electrical properties suggest the great potential of nanopaper FETs in next-generation of flexible and transparent electronics and in a broad range of other cost-efficient and practical applications.

EXPERIMENTAL METHODS

Nanopaper Preparation and Characterization. First, 78 mg of TEMPO was subjected to ultrasonication in 75 mL of deionized water for 10 min to achieve a uniform solution, and then the TEMPO solution was combined with 514 mg of NaBr dissolved in 50 mL of deionized water. The TEMPO/NaBr mixture was added to 5 g of dry weight Kraft bleached softwood pulp suspended in 65 g of deionized water. The TEMPO-mediated oxidation of the cellulose slurry was initiated by adding 30 mL of 12% NaClO aqueous solution at room temperature. The pH was controlled to be 10.5 by adjusting with 0.5 mol/L sodium hydroxide solution. The pH was monitored every 20 min for 2 h. Once the TEMPO treatment was finished, the fibrous TEMPO-oxidized product was washed with water by Büchner filtration until white product was obtained. It was then dispersed into water at a concentration of 1 wt % and disintegrated by one pass through the microfluidizer M-110EH (Microfluidics Ind., USA). The obtained NFC dispersion was diluted with deionized water, followed by mixing at 500 rpm using an Ultra Turrax mixer (IKA, RW20 digital) for 10 min. The final NFC dispersion concentration was 0.2 wt %. The dispersion was degassed for 20 min with a bath sonicator until no bubble was observed in the suspension. The dispersion was filtered by nitrocellulose ester filter membrane (Millipore DAWP29325) with 0.65 μm pore size. After filtration, a gel "cake" formed on top of the filter membrane. The "cake" was then placed between two smooth substrates and placed in an oven set at 40 °C for 10−15 min and then it was hot-pressed at 105 °C for 10 to 15 min. After drying, a transparent, flexible, and strong nanopaper with 90 mm diameter was obtained. A multimode atomic force microscope (Veeco Instruments) with a high aspect ratio tip was used to characterize the surface of nanopaper in tapping mode.

Transistor Fabrication. n-Type organic semiconductor NTCDI-F15 was synthesized according to published procedures.[22] Bottom-gate top-contact OFETs were fabricated on nanopaper substrates. SWCNT film was deposited on nanopaper by Meyer Rod coating method and then dried in the oven to form transparent and conductive gate electrodes. One μm of PMMA dielectric layer was deposited by spin-coating and then annealed at 90 °C for 15 min. Then 100 nm of n-type organic semiconductor NTCDI-F15 was vacuum deposited ($P \approx 2 \times 10^{-6}$ Torr) onto the dielectric layer with the substrate kept nominally at room temperature. Finally, silver electrodes were thermal evaporated through shadow masks with a channel length and width of 100 and 2000 μm, respectively. Two Keithley 2400 source meters were used to carry out electrical measurements. All nanopaper field-effect transistors were tested under ambient atmospheric conditions.

Conflict of Interest: The authors declare no competing financial interest.

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Supporting Information Available: SEM image of AZO by ALD on nanopaper surface; conductive CNT patterns drawn by a ballpoint pen on nanopaper. This material is available free of charge via the Internet at http://pubs.acs.org.

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