Supplementary information

Transparent, flexible and recyclable nanopaper-based touch sensors fabricated via inkjet-printing

Hao Ling, Ruwei Chen, Quanbo Huang, Feng Shen, Yuyuan Wang, Xiaohui Wang *

State Key Laboratory of Pulp & Paper Engineering, South China University of Technology, Guangzhou 510640, China.

1. AFM images of CNFs and SEM images of CNFs paper

Figure S1(a) shows the high-resolution atomic force microscopy (AFM) height images of cellulose nanofibrils. The water dispersion of cellulose nanofibrils with a mass fraction of 1% was diluted 1000 times with deionized water, and then put a drop on the mica sheet. Figure S1(b) shows the SEM images of cellulose nanofibrils paper.

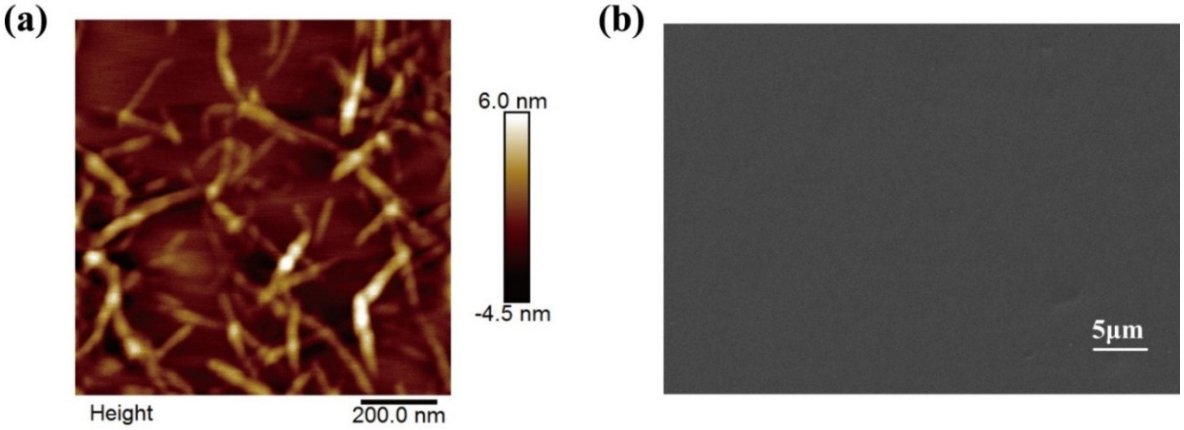

Figure S1. **(**a)Topographic (height) images of cellulose nanofibrils obtained with tapping-mode AFM. All images have a size of 1 μm × 1 μm; (b) SEM images of cellulose nanofibrils paper.

2. The transmittance of nanopaper and the NPTS

Figure S2 shows the transmittance of nanopaper, NPTS, annealed NPTS and annealed NPTS with DMSO treatment. The obtained nanopaper expresses a high optical transmittance of 89% at 640nm. After inkjet-printing PEDOT:PSS electrodes, the transmittance decreased to 82% at 430nm. The transmittance has hardly changed after annealing treatment. After DMSO treatment, the transmittance decreased slightly but still reached to 80% at 400nm.

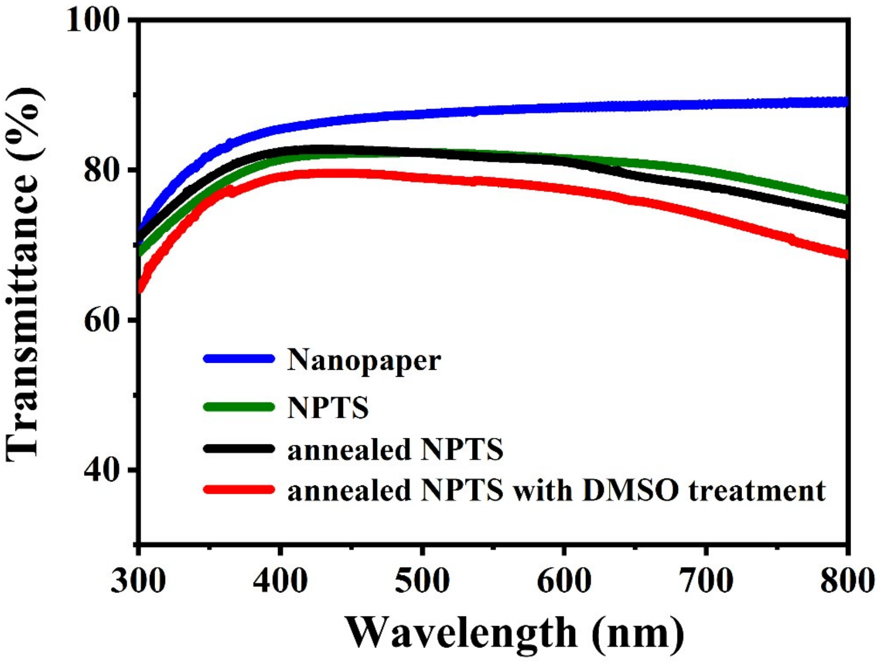

Figure S2. The transmittance of CNFs paper and the NPTS.

3. Printing process of the NPTS

Figure S2(a) shows the image of the ME33 EPSON inkjet printer. Figure S2(b) shows the printing process of the NPTS. At first, the PEDOT:PSS pattern was printed on the ordinary print paper to mark the print area. Afterward, cellulose nanofibrils paper was covered and fixed on this area. By performing the printing process 6 times and then annealing at 80 ℃ for 1 h, the NPTS with a single-keypad was prepared.

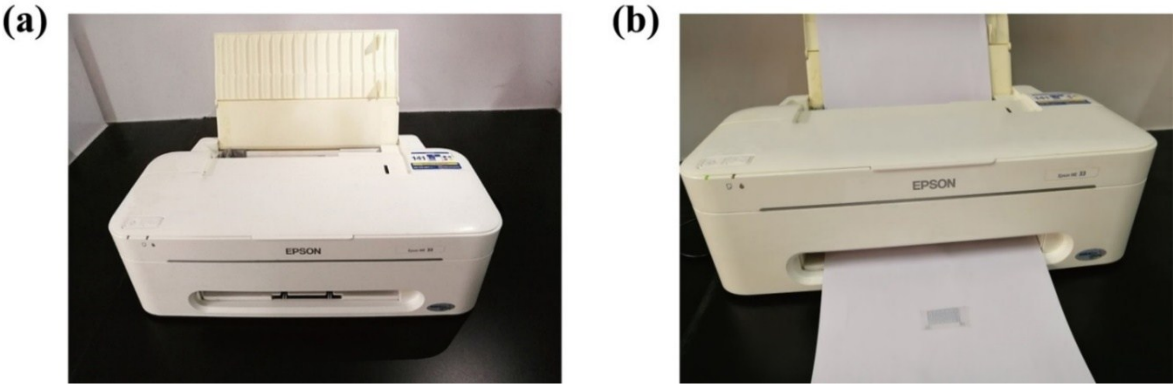

Figure S3. (a) Image of ME33 EPSON inkjet printer; (b) Printing process of the NPTS.

4. Materials cost calculation

The cellulose nanofibrils used in this experiment were made by our laboratory. The market price was used in the cost calculation for facilitating calculation.

Table S1. Materials cost calculation

Materials cost of a NPTS (0.0465g, 3.5 cm × 2.5 cm × 0.05 mm)			
Materials	Quality (g)	Price ($/g)	Cost ($)
cellulose nanofibrils	0.0393	1.177	0.046256
PEDOT:PSS	0.0071	1.1299	0.008022
p-toluenesulfonic acid	5.32×10^{-5}	0.8362	0.000044
DMSO	0.11	0.05085	0.005594
Total price			0.059916

5. The stress-strain curve of different paper substrates.

The tensile strength of the NPTS is 104.23Mpa which is equal to that of nanopaper, due to the great stretchability of PEDOT:PSS electrodes.[1] During the tensile test, the PEDOT:PSS electrodes will not be damaged unless the nanopaper breaks.

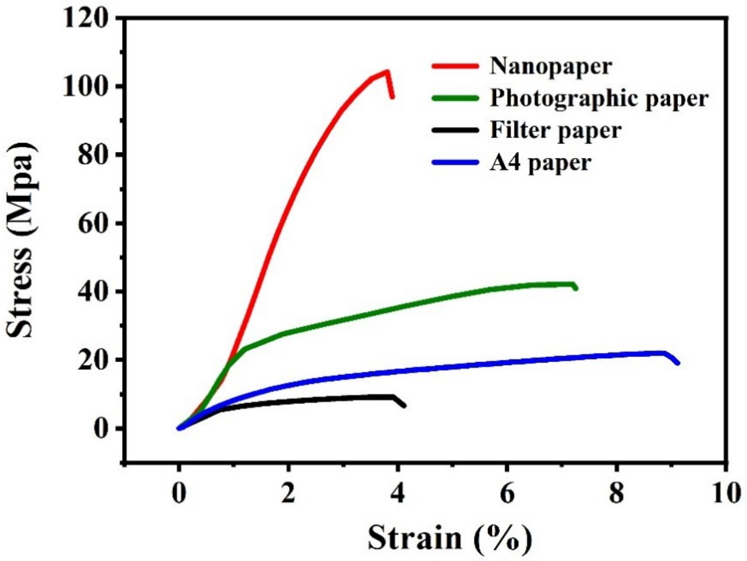

Figure S4. The stress-strain curve of different paper substrates.

6. Chemical Structure of PEDOT:PSS

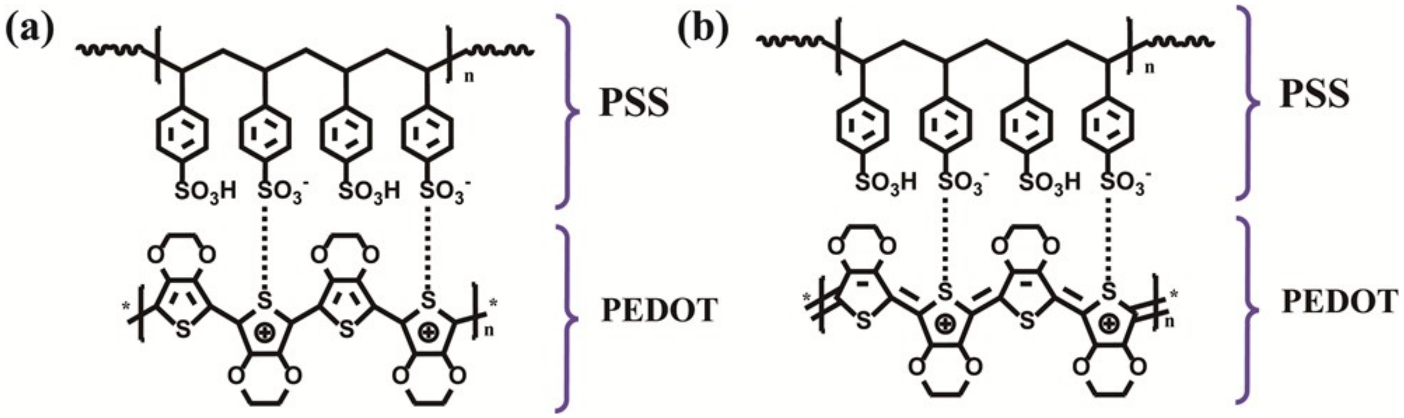

Figure S5. Different structure of PEDOT:PSS. (a) Benzoid structure; (b) Quinoid structure.

7. 3D topographic images of PEDOT:PSS films obtained with tapping-mode AFM

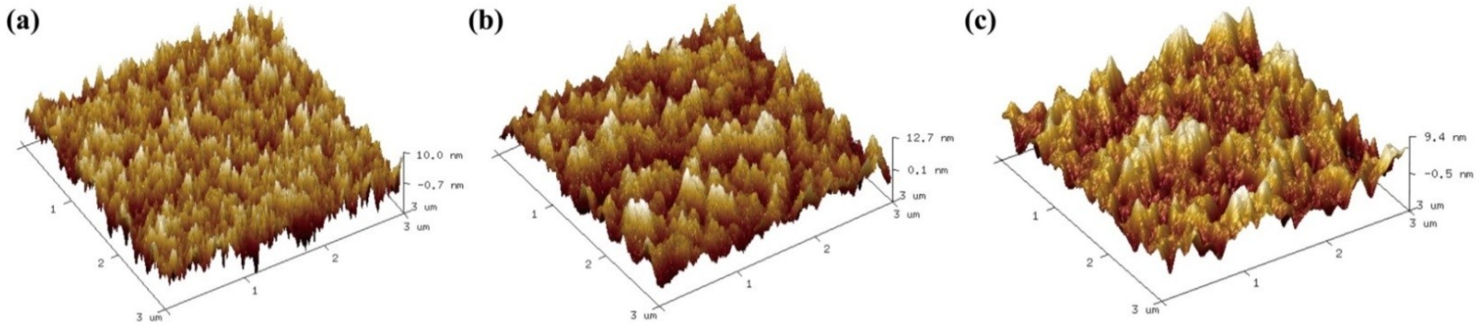

Figure S6. 3D topographic images of PEDOT:PSS film obtained with tapping-mode AFM. (a) The pristine PEDOT:PSS film; (b) The PTSA doped PEDOT:PSS film; (c) The PTSA doped PEDOT:PSS film with DMSO post-treatment. All images have a size of 3 μm × 3 μm.

8. Annealing effect

After annealing in the vacuum oven for 1 h under 80 ℃, the conductivity of PEDOT:PSS can be increased by 40%~70%. The reason is that annealing decreased the surface roughness, which reduced the defect density on the surface of the PEDOT:PSS film, thus effectively improving the conductivity of the film.[2]

Table S2. The variation of electrical conductivity of PEDOT:PSS before and after annealing

<table>
<tr><td colspan="7">The concentration of deionized water is 30 wt%</td></tr>
<tr><td>Concentration of PTSA (wt%)</td><td>0</td><td>0.25</td><td>0.50</td><td>0.75</td><td>1.00</td><td>1.25</td></tr>
<tr><td>Conductivity before annealing (S/cm)</td><td>19</td><td>56</td><td>195</td><td>456</td><td>593</td><td>780</td></tr>
<tr><td>Conductivity after annealing (S/cm)</td><td>32</td><td>90</td><td>290</td><td>690</td><td>920</td><td>1130</td></tr>
<tr><td>Conductivity increment</td><td>68.4%</td><td>60.7%</td><td>48.7%</td><td>51.3%</td><td>55.1%</td><td>44.9%</td></tr>
<tr><td colspan="7"></td></tr>
<tr><td colspan="7">The concentration of PTSA is 0.75 wt%</td></tr>
<tr><td>Concentration of water (wt%)</td><td>0</td><td>10</td><td>20</td><td>30</td><td colspan="2">40</td></tr>
<tr><td>Conductivity before annealing (S/m)</td><td>577</td><td>490</td><td>420</td><td>456</td><td colspan="2">356</td></tr>
<tr><td>Conductivity after annealing (S/m)</td><td>870</td><td>786</td><td>711</td><td>690</td><td colspan="2">590</td></tr>
<tr><td>Conductivity increment</td><td>50.7%</td><td>60.4%</td><td>69.3%</td><td>51.3%</td><td colspan="2">65.7%</td></tr>
</table>

9. Equivalent circuit of the NPTS in parallel with an oscilloscope probe

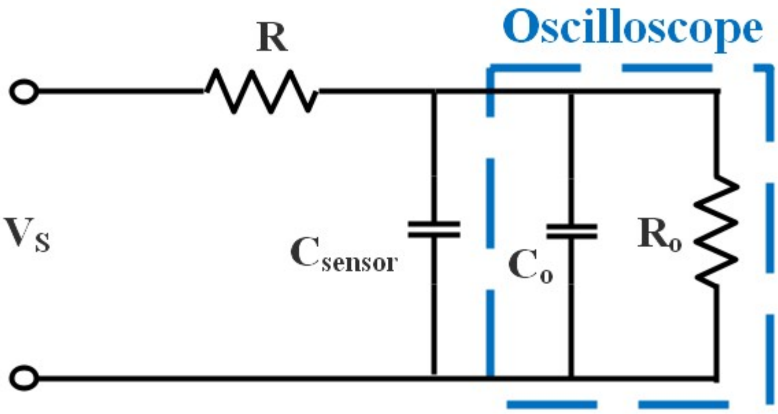

Figure S7. Equivalent circuit of the NPTS in parallel with an oscilloscope probe

10. Schematic diagram of the NPTS

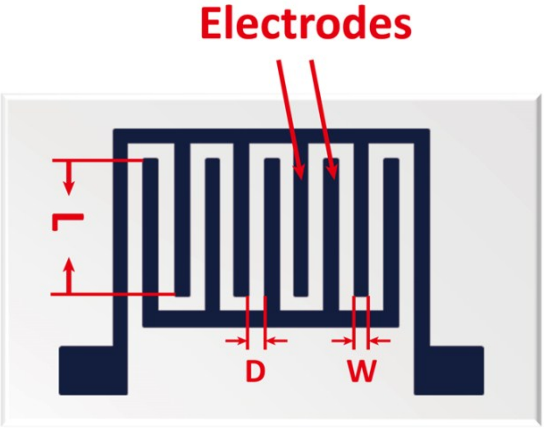

Figure S8. Schematic diagram of the NPTS.

11. Sealing effect

The NPTS without sealing showed diminished conductivity over time in high humidity due to the water absorption of PSS chains. As the PSS chains absorbed the water, the distance between the adjacent PEDOT chains increases. Consequently, the hopping conduction mechanisms of charges between PEDOT chains become harder and the conduction reduces.[3] After 5 hours, the conductivity tended to stabilize due to the saturated water absorption of PSS chains. Besides, the nanopaper substrate showed bending due to the water absorption of cellulose fibers.

After sealing the NPTS with a passivation layer of adhesive tape, the conductivity of the NPTS retained stable even after 3 days and the nanopaper substrate showed no bending.

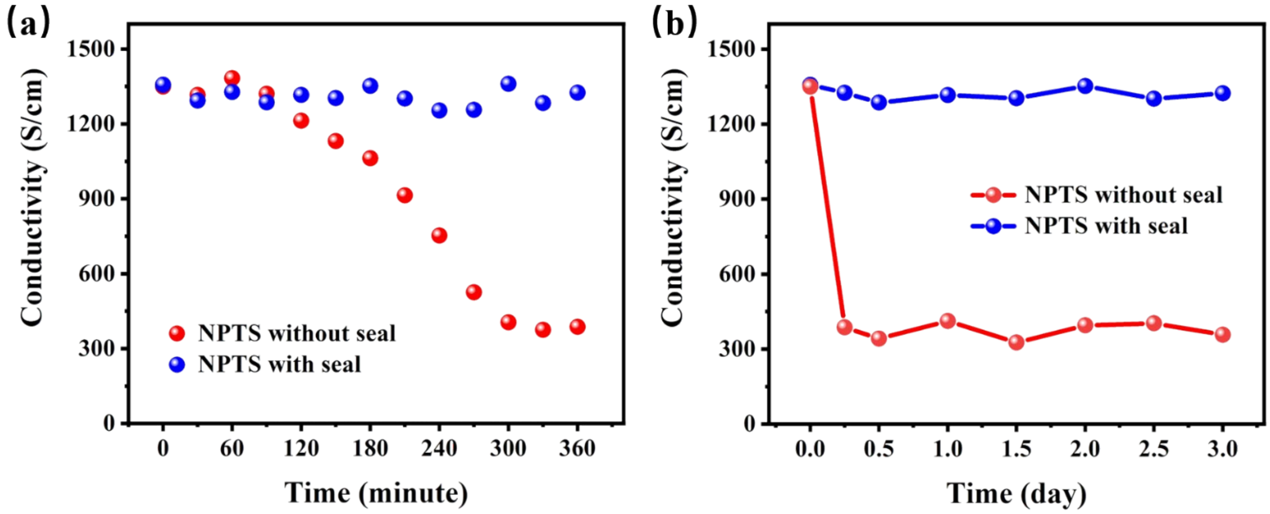

Figure S9. The variation of electrical conductivity of the NPTS at 80%RH with time.

12. Set of Arduino UNO

The I/O pin of Arduino UNO was connected to the active electrode as well as the GND pin was connected to the grounded electrode. At first, we set the I/O pin to output and the LOW state to discharge the capacitor which connects the pin. Afterward, the internal pull-up resistor (35.7 kΩ) was turned to the HIGH state, measuring the time required to return to the HIGH state. The pull-up resistor integrated into the microcontroller cascaded with the touch sensor capacitor to serve as an “RC-circuit”. The elapsed time is defined as the time required for the potential rise from the low value to a defined value (3V for Arduino UNO), which is in proportion to capacitance. Set the frequency of discharging-pull-up cycles to 1 kHz.

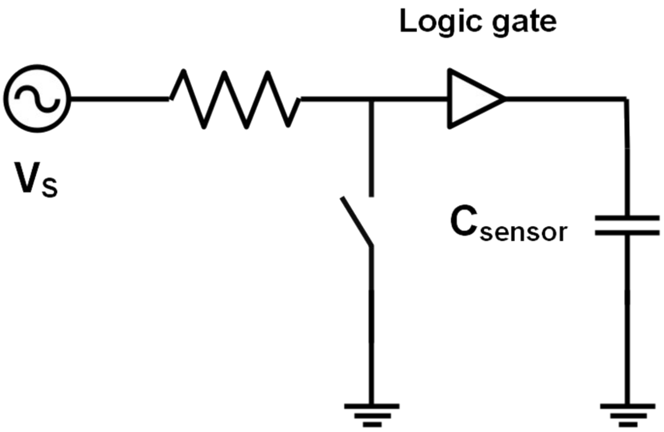

Figure S10. Equivalent circuit of internal pull-up circuits.

13. The threshold for Arduino UNO

To receive signals from the NPTS, a time threshold was set to make a distinction between the touched and untouched state. In the initial state, the elapsed time is below the threshold and the keypad is defined as untouched. Once the elapsed time exceeds this threshold, the keypad is defined to be in a touched state. The elapsed time for Arduino UNO is calculated as in Figure S11.

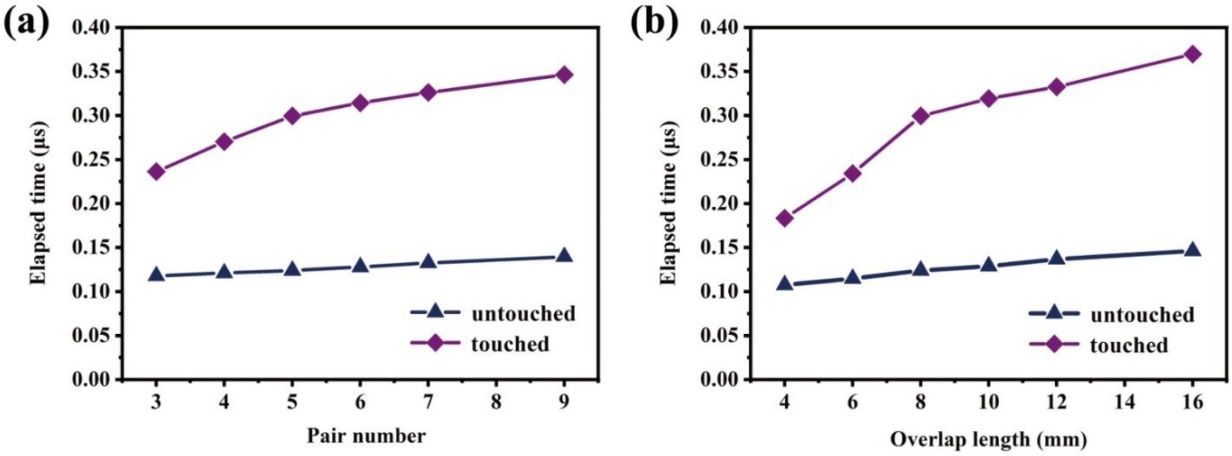

Figure S11. The elapsed time of the NPTS with (a) different numbers of microelectrodes digits and (b) different overlapping lengths of microelectrodes digits in untouched and touched case.

Video

Demonstration of the NPTS with four keypads.

Supplementary References

1. D. J. Lipomi, J. A. Lee, M. Vosgueritchian, B. C.-K. Tee, J. A. Bolander and Z. Bao, *Chemistry of Materials*, 2012, **24**, 373-382.
2. Z. Xiong and C. Liu, *Organic Electronics*, 2012, **13**, 1532-1540.
3. M. Kuş and S. Okur, *Sensors and Actuators B: Chemical*, 2009, **143**, 177-181.