Supporting Information

Inkjet printing of homogeneous and green cellulose nanofibrils delectric for high performance IGZO TFTs

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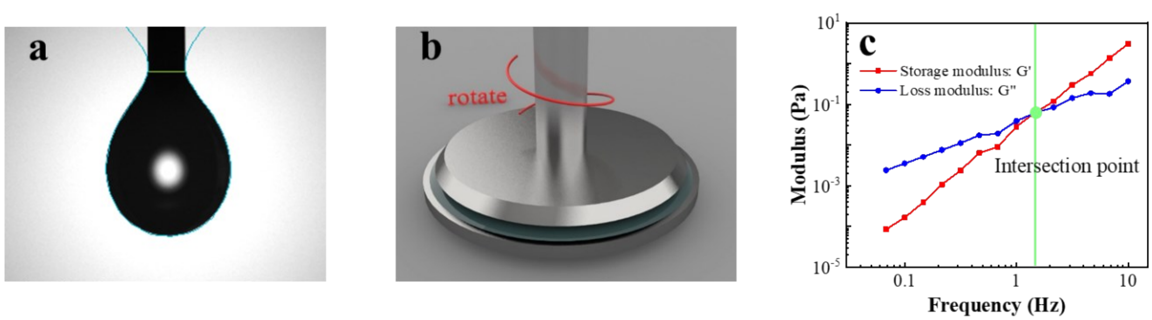

Figure S1. (a) Surface tension measured by pendant drop method. (b) Viscosity measured by HAAKE MARS 40 Rheometer. (c) Viscoelasticity measured by HAAKE MARS 40 Rheometer.

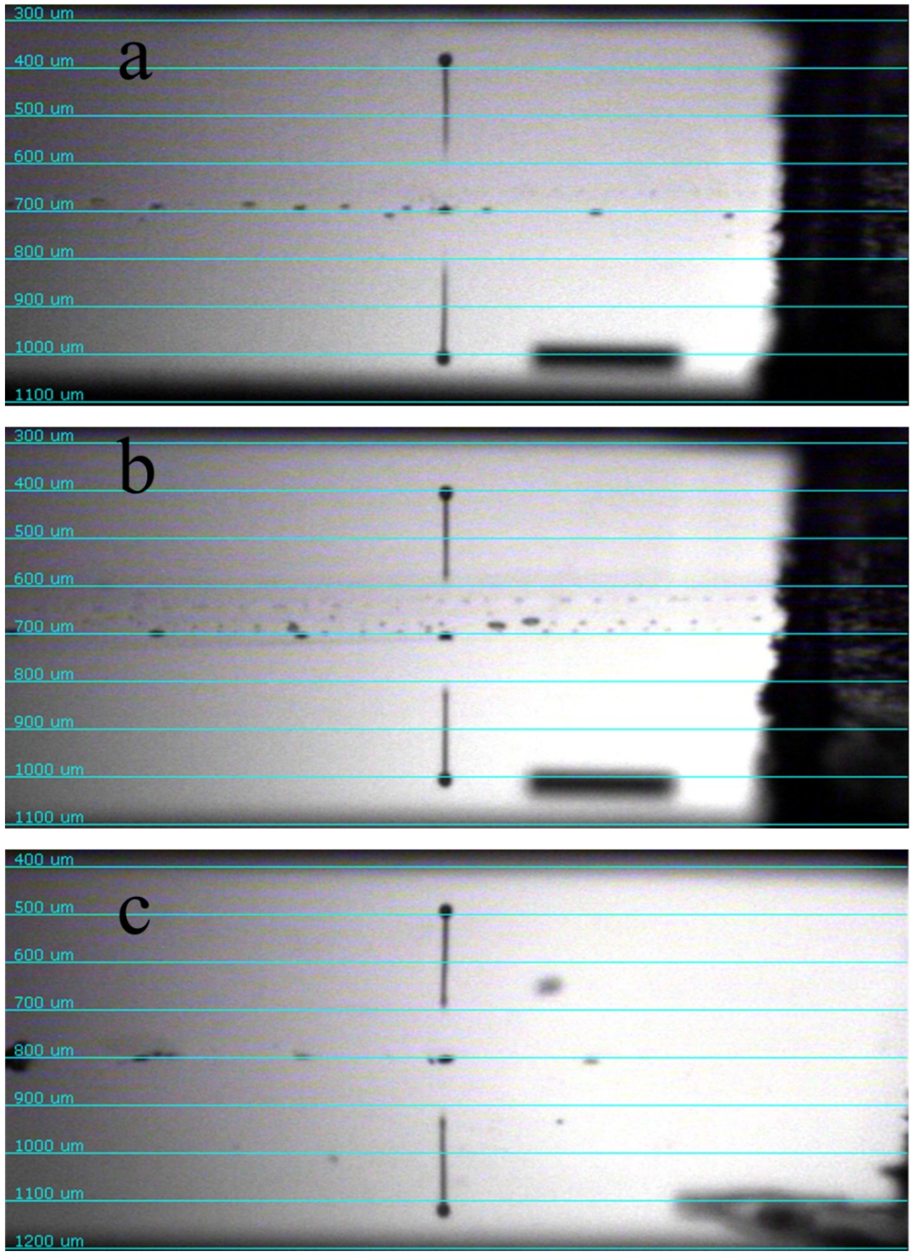

Figure S2. (a) Initially droplet ejection of the CNF ink No. 7. (b) Droplet ejection of the CNF ink No. 7 after printing for 1 hours. (c) Droplet ejection of the CNF ink No. 7 after 3 months storage.

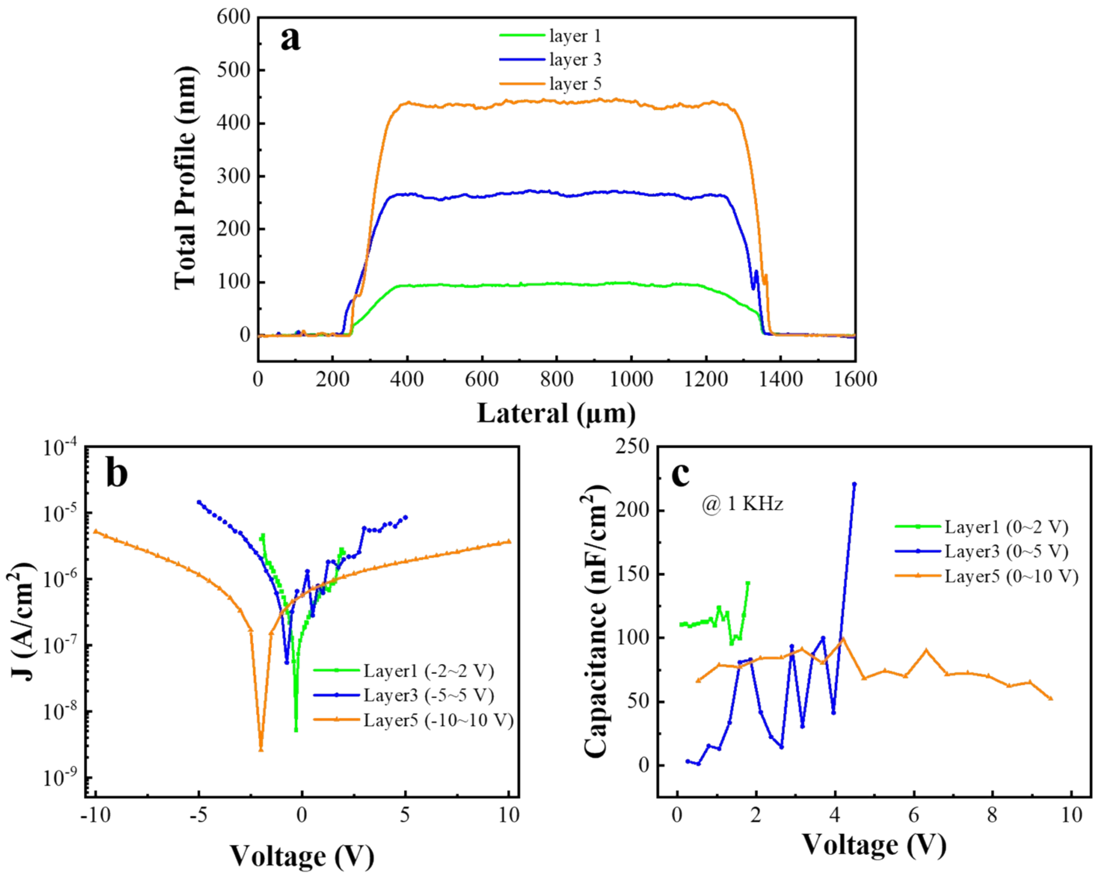

Figure S3. The dielectric properties of printed CNF with different thicknesses.

It can be seen that the film thickness increased with the increase of printing layers. The 5-layer printed CNF film, with a thickness of about 450 nm, showed the lowest leakage current and a stable capacitance. For 1-layer and 3-layer printed films, the leakage current was relatively large and the capacitance fluctuated with the voltage.

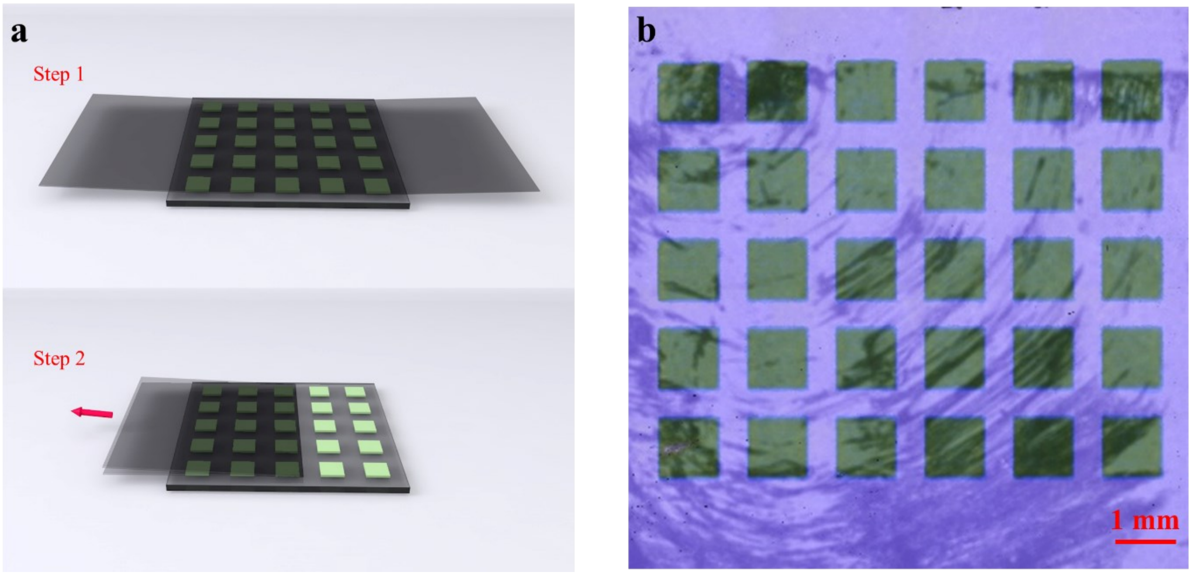

Figure. S4. (a) The procedure of tape test. (b) The photograph of the printed CNF film on indium-tin-oxide (ITO) substrate after tape test.

Firstly, the tape was placed over the printed CNF films by a finger. To ensure good contact with the film rubbed the tape firmly with the eraser, and the color under the tape is a useful indication of when good contact has been made. Generally, the adhesion strength of the tape with the film has been determined when good contact was established. Within 90 ± 30 s of application, the tape was removed at as close to an angle of 180° as possible. Rate the adhesion between film and substrate in accordance with the following scale:

5B) The edges of the cuts are completely smooth; none of the squares of the lattice is detached.

4B) Small flakes of the coating are detached at intersections; less than 5 % of the area is affected.

3B) Small flakes of the coating are detached along edges and at intersections of cuts. The area affected is 5 to 15 % of the lattice.

2B) The coating has flaked along the edges and on parts of the squares. The area affected is 15 to 35 % of the lattice.

1B) The coating has flaked along the edges of cuts in large ribbons and whole squares have detached. The area affected is 35 to 65 % of the lattice.

0B) Flaking and detachment worse than Grade 1.

Fig. S4 (b) is the photograph of the printed CNF film on indium-tin-oxide (ITO) substrate after tape test. It can be seen that almost none of the CNF film squares was detached and the edges of the CNF film squares were completely smooth. Thereby, the adhesion of printed CNF films with substrate was rated as 5B.

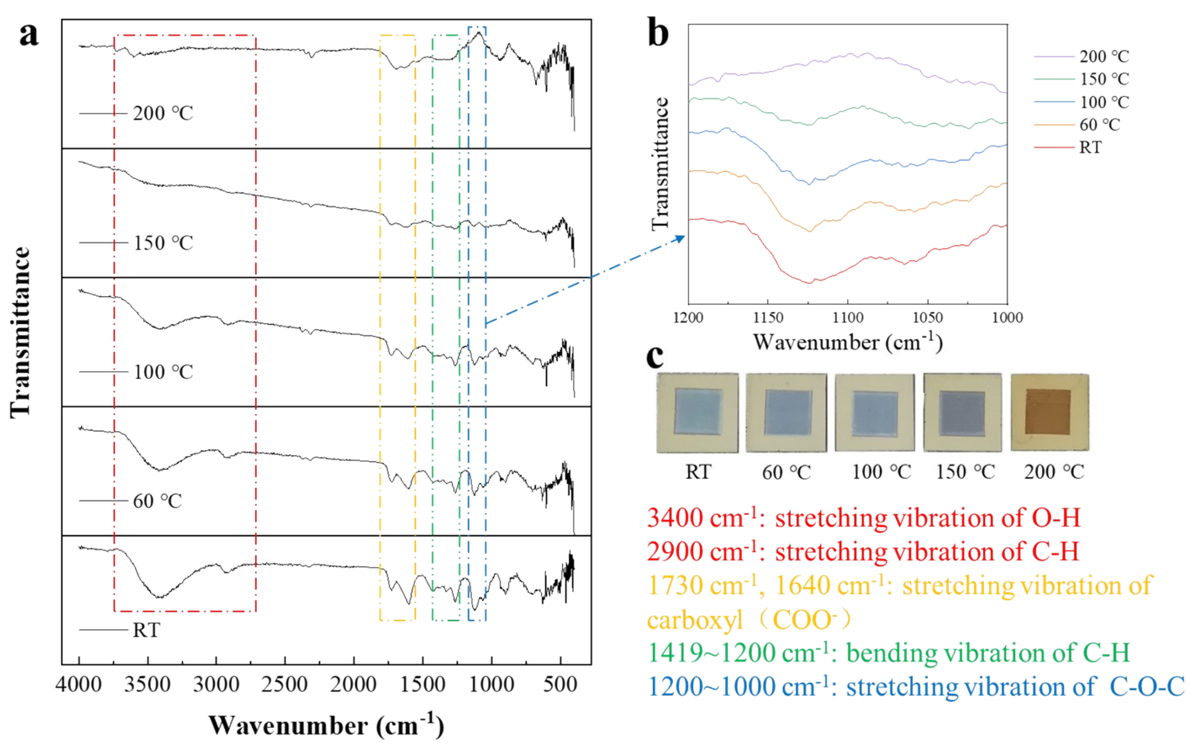

Figure. S5. FTIR result of printed NFC dielectrics with different annealing temperatures (from room temperature to 200 °C).

Fourier transform infrared (FTIR) spectroscopy measurement was carried out for printed NFC dielectrics with different annealing temperatures. The broad band absorption centered around 3400 cm^{-1} is ascribed to the stretching of stretching vibration of O–H, while the narrow band absorption located at 2900 cm^{-1} can be associated with stretching vibration of C–H group. The double absorption peaks at 1730 and 1640 cm^{-1} are assigned to stretching vibration of carboxyl (COO^-), and peaks at 1200~1000 cm^{-1} are attributed to the stretching vibration and bending absorption of C-O-C group. It can be seen from the Fig. S5 (b) that as the temperature rose from room temperature (RT) to 200 °C, the intensity of the C-O-C absorption peak gradually decreased and finally disappeared at 200 °C. Correspondingly, the printed CNF film annealed at 200 °C degraded and turned yellow as shown in Fig. S5 (c). In conclusion, the printed CNF dielectric layer can exist stably at a temperature below 150°C.

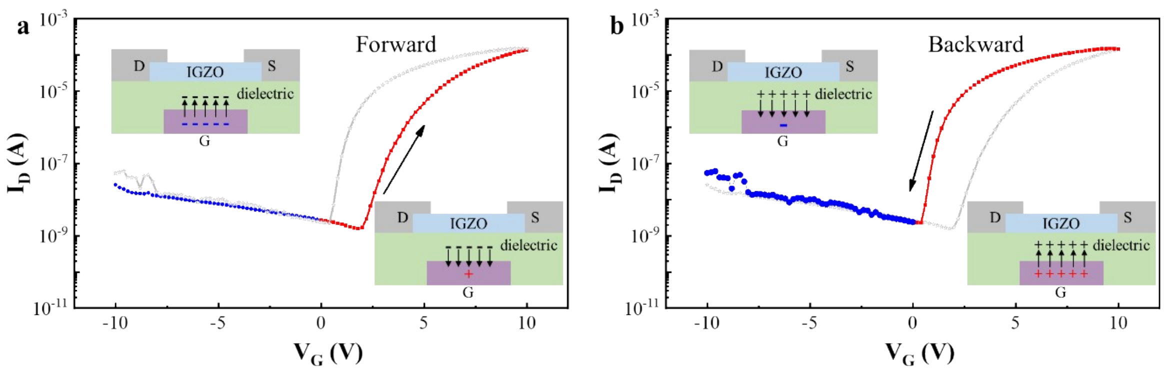

Figure. S6 The possible mechanism of hysteresis phenomena.

When V_G is swept from negative to positive value (Forward), the electrons are injected into the dielectric layer from the gate and trapped by hydroxide groups. After the gate voltage changes from negative to positive, part of the voltage is used to remove the bound carriers, which will be mainly responsible for the positive shift of the transfer curve. Similarly, when V_G is swept from positive to negative value (Backward), the holes can be injected from the gate and trapped by hydroxide groups, resulting in a negative shift of the transfer curve. For both cases, the charges injected from the gate are considered to be the crucial factor causing hysteresis.

Table S1. Element types and contents in the printed CNF film measured by XPS

Name	Atomic %
C 1s	58.68
O 1s	34.84
N 1s	0.17
Cl 2p	0.06
Na 1s	5.46
F 1s	0.8