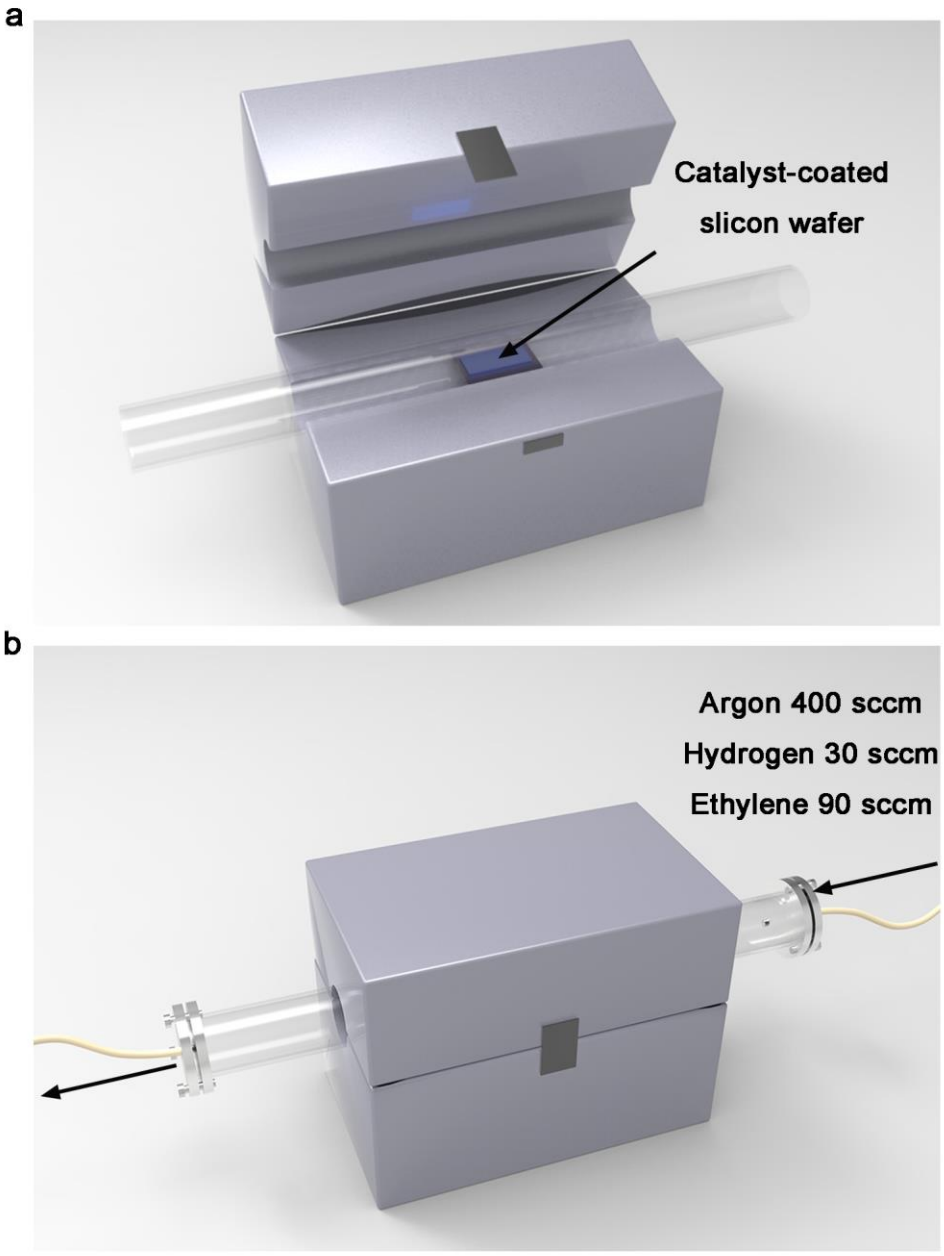
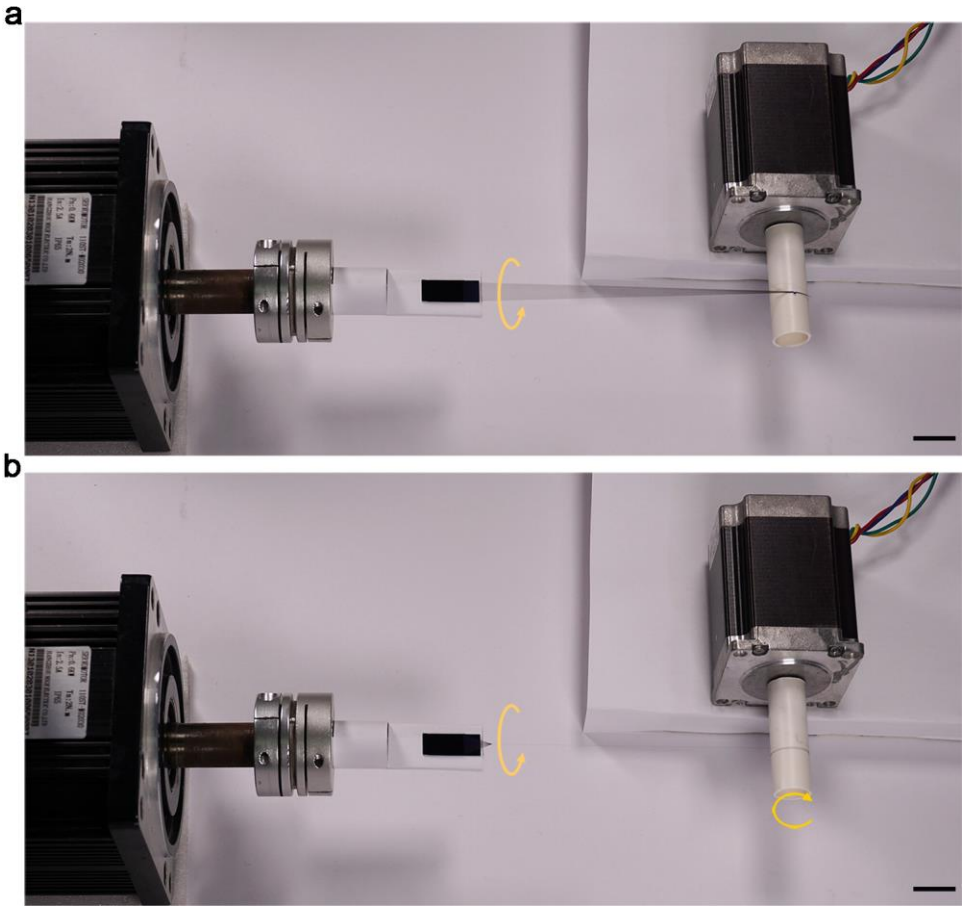


Supplementary Figure 1

Growth of CNT arrays through CVD process by a tube furnace.

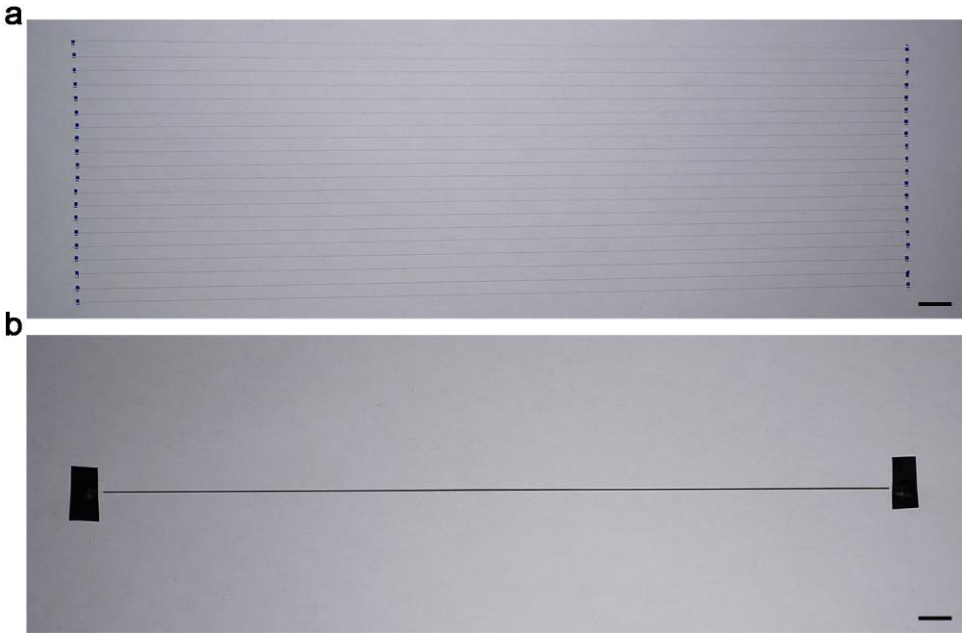
(**a**) Placing the catalyst-coated silicon wafer in the middle of the tube reactor. (**b**) Schematic illustration to the growth of spinnable CNT array.



Supplementary Figure 2

Preparation of the primary CNT fibre.

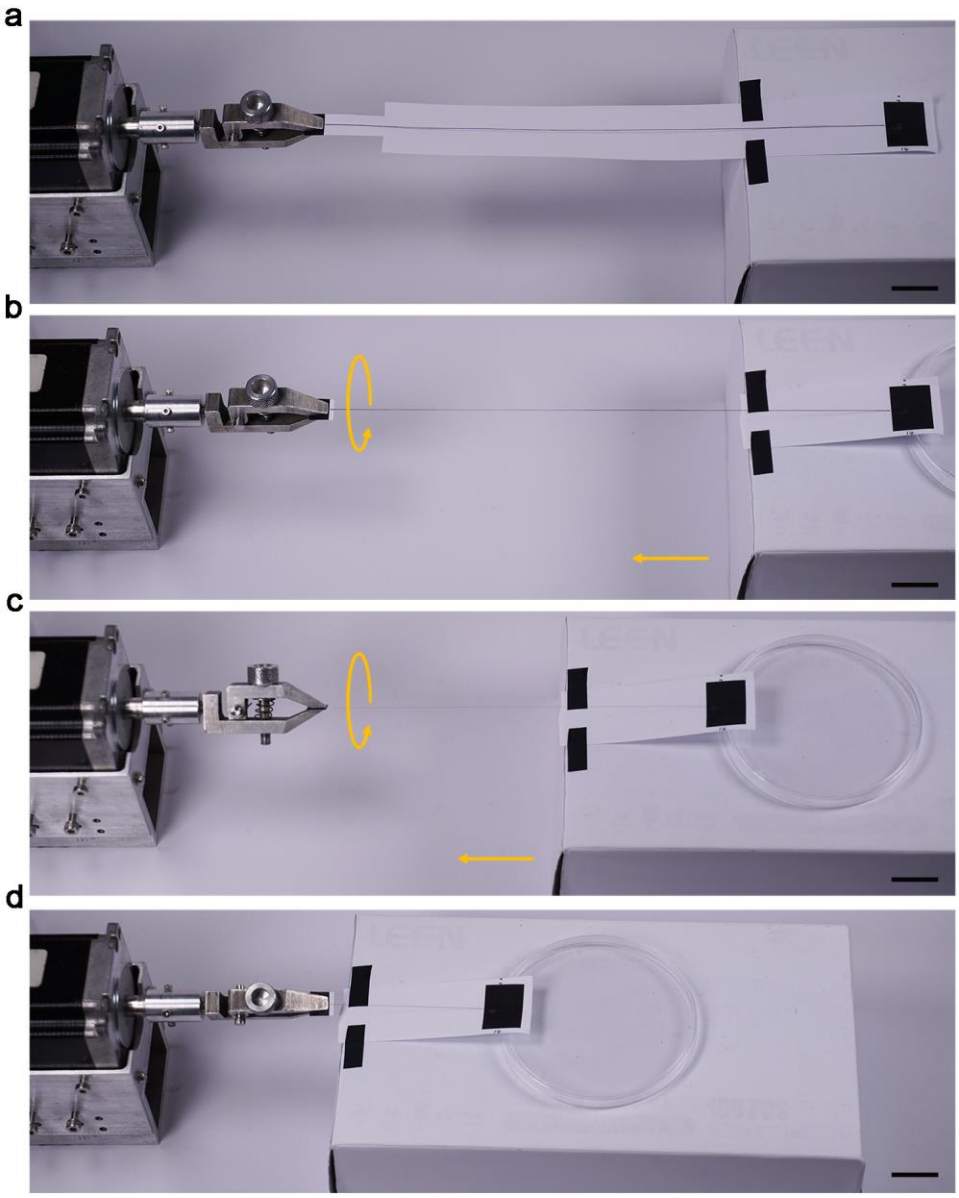
(**a**) Placing a CNT array on the rotor of a serve motor (left) with the end of the spun CNT sheet on a collecting drum of a stepping motor (right). (**b**) Twisting the primary CNT fibre by anticlockwise rotation of the servo motor and clockwise rotation of the stepping motor. Scale bar in **a** and **b**, 2 cm.



Supplementary Figure 3

Arrangement of the multi-ply primary CNT fibres.

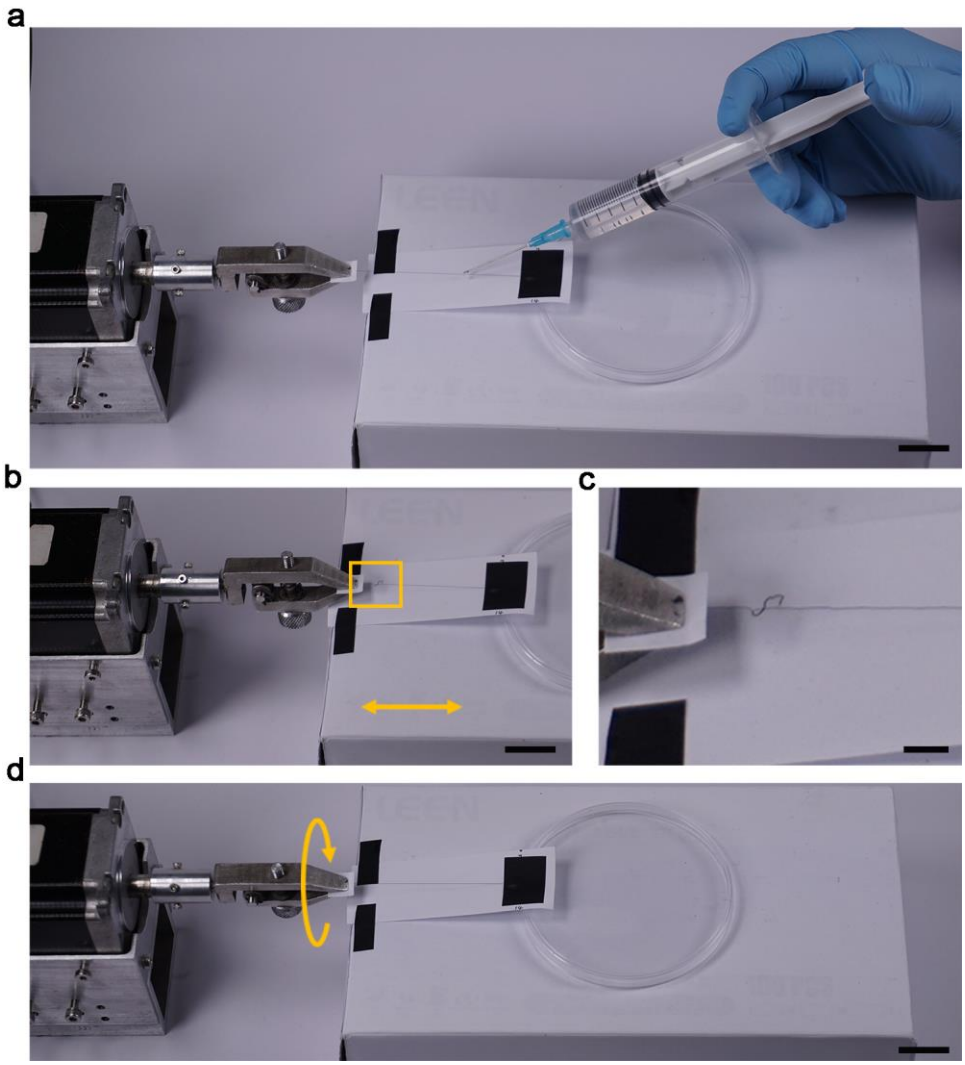
(**a**) Twenty parallel primary fibres with the same length of 25 cm on a sheet of paper. (**b**) Assembling the twenty parallel primary fibres together. Scale bar in **a** and **b**, 1 cm.



Supplementary Figure 4

Preparation of an HHF.

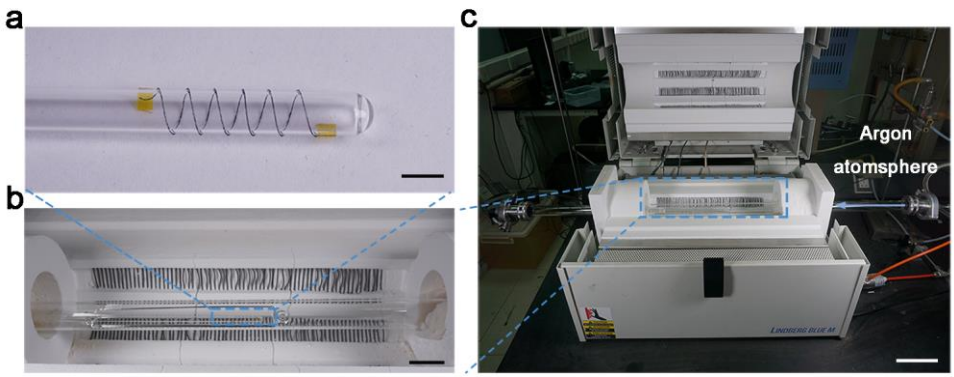
(**a**) Clamping one end of the 20-ply fibre onto a rotating motor shaft. The other end is attached to a moveable paper, and the length of the fibre is 25 cm. The fibre lays on the top of the paper. (**b**) Cutting the left part of the paper. (**c**) Anticlockwise twisting of the CNT fibre with the moveable end being moved towards the shaft while keeping the fibre horizontal. (**d**) The completion of a coiled fibre. Scale bar in **a**-**d**, 2 cm.



Supplementary Figure 5

Post-treatment of an HHF.

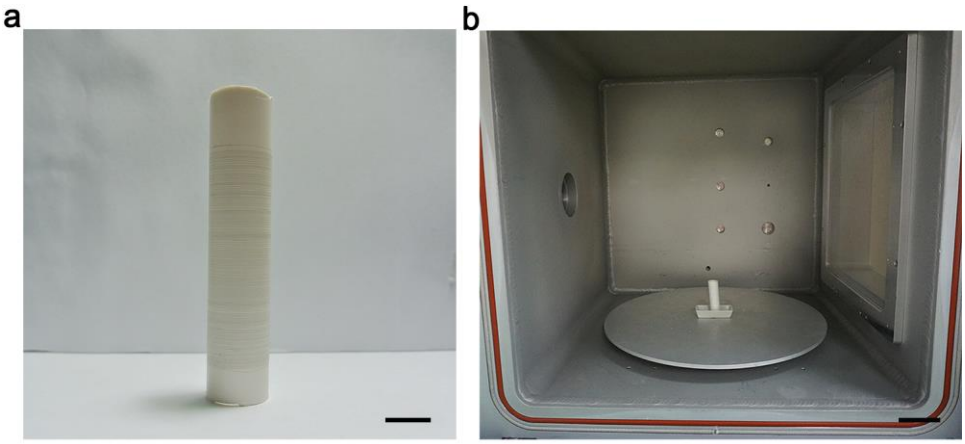
(**a**) Infiltrating the whole fibre with ethanol. (**b**) Slightly relaxing the fibre to observe the excessive twisting by moving the moveable substrate towards the motor. (**c**) An entanglement confirming the excessive twisting. (**d**) Clockwise rotation of the motor to release the excessive twisting. Scale bar in **a**, **b** and **d**, 2 cm. Scale bar in **c**, 2 mm.



Supplementary Figure 6

The preparation of a spring by a thermo-hydro setting of an HHF.

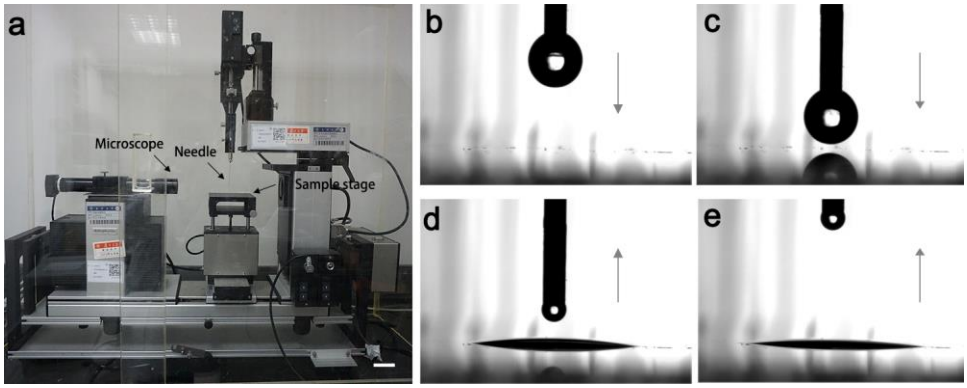
(**a**) Spirally wrapping an HHF on a glass rod. Scale bar, 5 mm. (**b**) The glass rod being transferred into a quartz boat at the middle of a furnace tube. Scale bar, 3 cm. (**c**) The HHF being heated at 300 ^{o}C for 1 h in the furnace at argon atmosphere (it is heated to 300 ^{o}C in 15 min). Scale bar, 10 cm.



Supplementary Figure 7

The preparation of hydrophilic fibres.

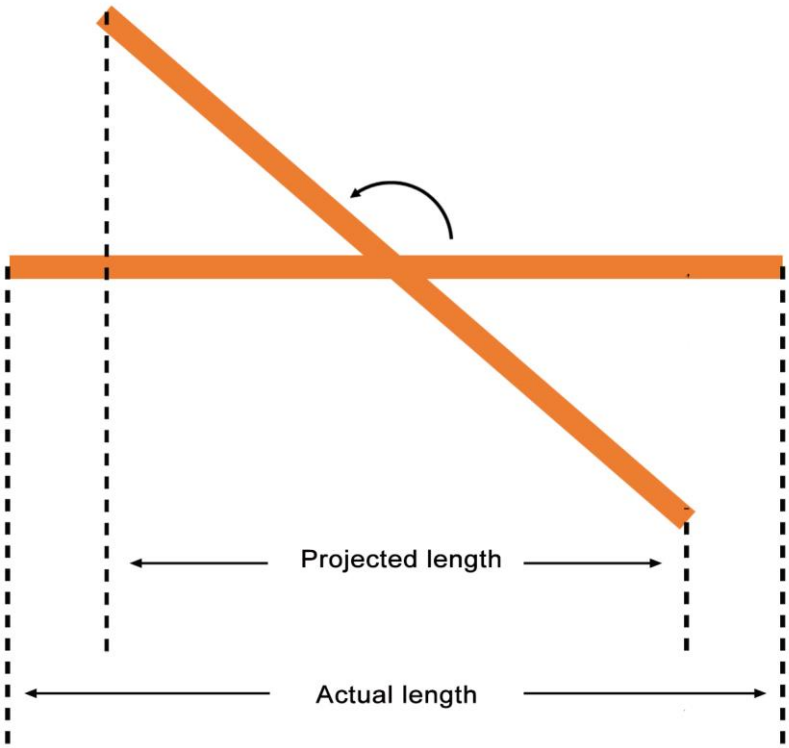
(**a**) Collecting primary fibres on a drum with arrangement at regular intervals. Scale bar, 1 cm. (**b**) Modifying the fibre under oxygen microwave plasma. Scale bar, 5 cm.



Supplementary Figure 8

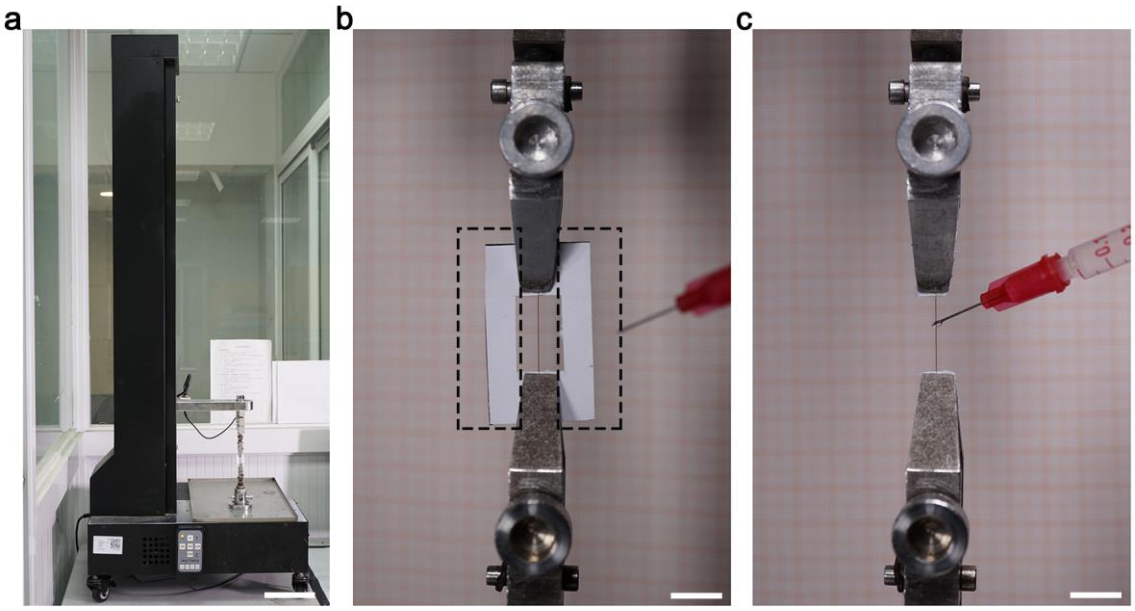
The measurement of contact angle.

(**a**) Experimental setup for the contact angle measurement. Scale bar, 3 cm. (**b**-**e**) The detailed process of a water droplet comes into contact with the sample.



Supplementary Figure 9

Schematic illustration to the actual and projected lengths of the copper paddle for calculating the rotation angle.



Supplementary Figure 10

Contractive measurements.

(**a**) Experimental setup for the contractive measurement. Scale bar, 10 cm. (**b**) Clamping the two ends of the fibre on the table-top universal testing instrument. Scale bar, 1 cm. (**c**) Cutting the two shoulders of the paper (the area marked in dashed line in **b**), followed by touching with the solvent. Scale bar, 1 cm.