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 °C for 1 h in the furnace at argon atmosphere (it is heated to 300 °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.