Supporting Information

1. Two Methods to Fabricate Composite CNT/Silica Fibers

(1) Direct casting of the silicate precursor on the outer surface of CNTs by spinning pure CNT fibers through precursor solutions, followed by hydrolysis and condensation reactions of precursors in acid solutions (Figure S1).

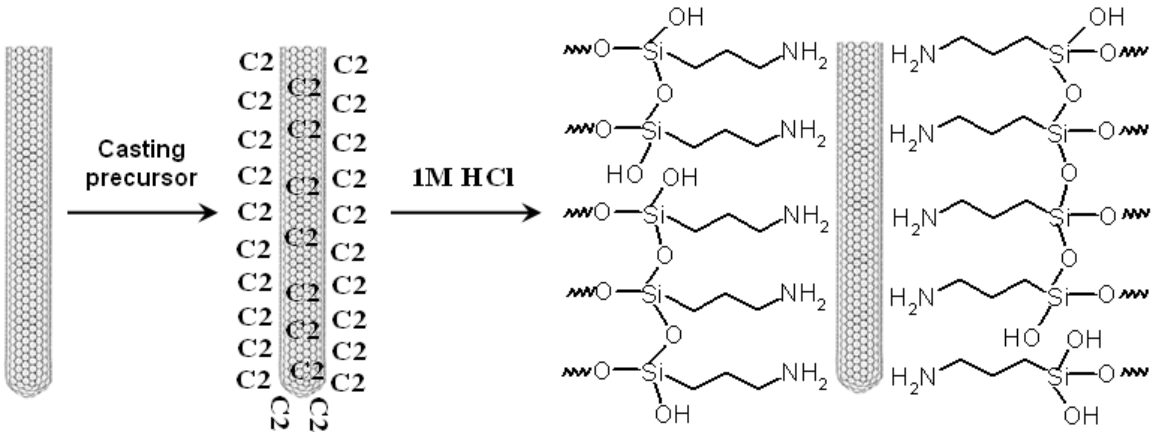

Figure S1. Schematic illustration of the synthesis of C2-derived composite CNT/silica fibers by direct casting.

(2) Chemical modification of the outer surface of CNTs (Figure S2). CNT fibers are first treated with 5 M HNO_3 to produce functional carboxylic acid groups on the outer surface of CNTs. C2 molecules are then connected to CNTs by hydrogen bonding. Following hydrolysis and condensation reactions of precursors in acid solutions produce composite CNT/silica fibers.

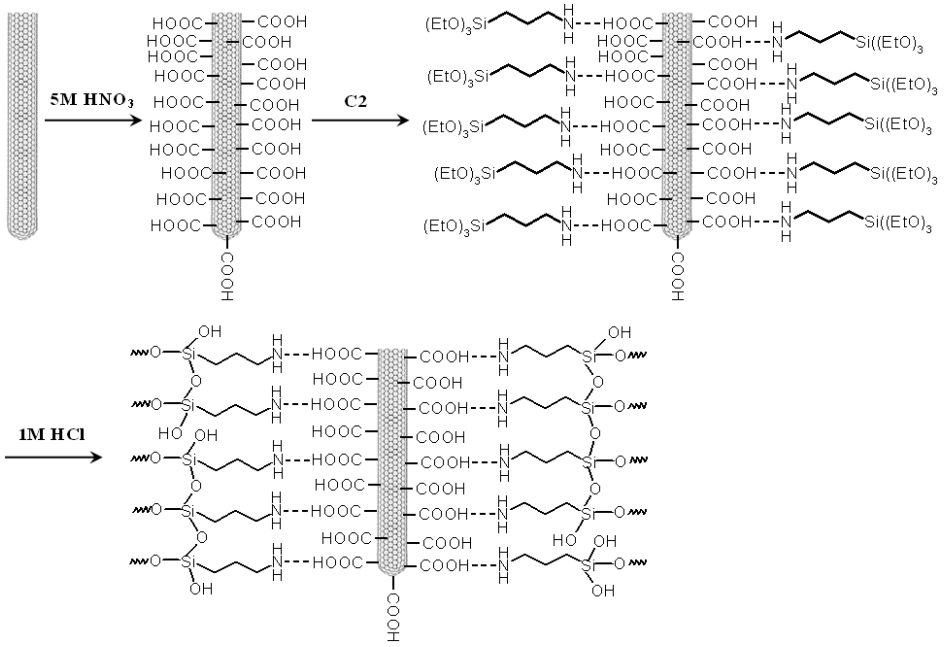

Figure S2. Schematic illustration of the synthesis of C2-derived composite CNT/silica fibers by chemical modification.

2. Calculation of the SiO_2 contents in the composite CNT/silica fibers

Samples 1, 2, 3, and 4 correspond to the CNT/silica fiber derived from C1 by direct casting, the CNT/silica fiber derived from C2 by direct casting, the pure CNT fiber treated by HNO_3, and the CNT/silica fiber derived from sample 3 followed by connection of C2 through chemical modification, respectively.

Table S1. The molar and weight contents of Si, C, and O.

Sample	Si (mol %)	Si (wt %)	O (mol %)	O (wt %)	C (mol %)	C (wt %)
1	1.69	3.81	4.49	5.77	93.82	90.42
2	2.13	4.78	3.86	4.94	94.01	90.28
3	0	0	9.99	12.88	90.01	87.12
4	4.50	9.50	16.60	19.28	78.90	71.22

For Sample 1:

The weight content of SiO_2 is calculated as $(1.69 \times 28.09 + 3.38 \times 16.00)/(1.69 \times 28.09 + 4.49 \times 16.00 + 93.82 \times 12.01) = 101.55/1246.09 = 8.15\%$. Here 28.09, 16.00, and 12.01 are the atomic mass of Si, O, and C, respectively.

For Sample 2:

The weight content of SiO_2 is calculated as $(3.86/2 \times 28.09 + 3.86 \times 16.00)/(2.13 \times 28.09 + 3.86 \times 16.00 + 94.01 \times 12.01) = 115.97/1250.65 = 9.27\%$.

For Sample 4:

The content of O in the calculation of SiO_2 comes from C2, not including the COOH groups derived from the oxidization by HNO_3. The calculation is detailed as below. The molar ratio between O and C in Sample 3 is $9.99/90.01 = 0.113$, so the O molar content from COOH groups in Sample 4 is $0.111 \times 78.90 = 8.75$. Therefore, the O molar content from C2 is $(16.60 - 8.75) = 7.85$. The weight content of SiO_2 is decided by $(7.85/2 \times 28.09 + 7.85 \times 16.00)/(4.50 \times 28.09 + 16.60 \times 16.00 + 78.90 \times 12.01) = 235.85/1330.60 = 17.73\%$.