

# Electronic Supplementary Materials

**Flexible All-Carbon Nanoarchitecture Built from In Situ**

**Formation of Nanoporous Graphene Within “Skeletal-Capillary”**

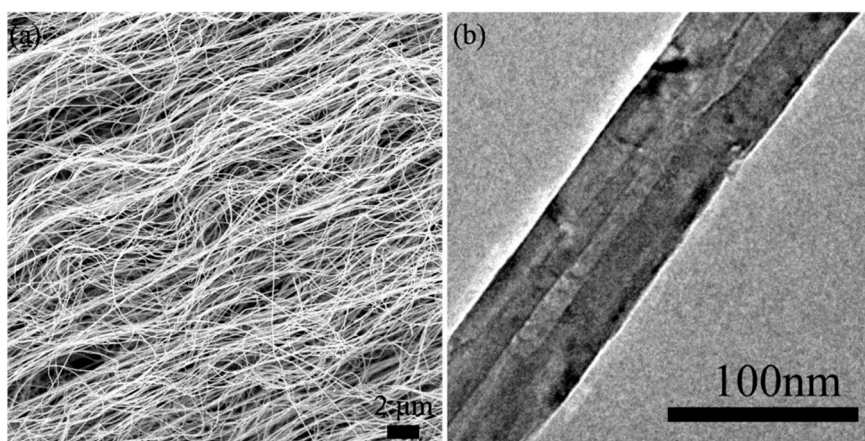
**Carbon Nanotube Networks for Supercapacitors**

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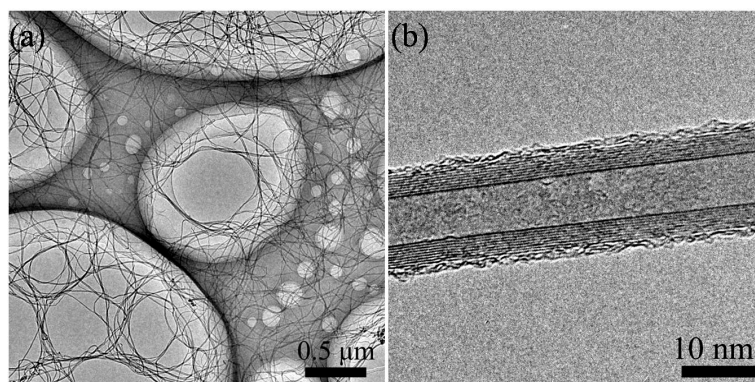
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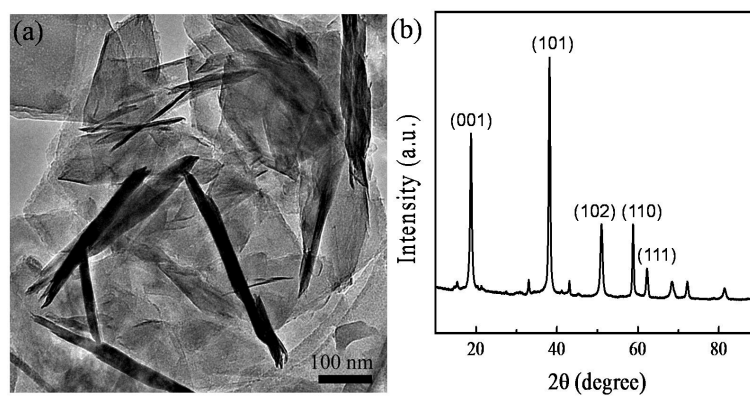
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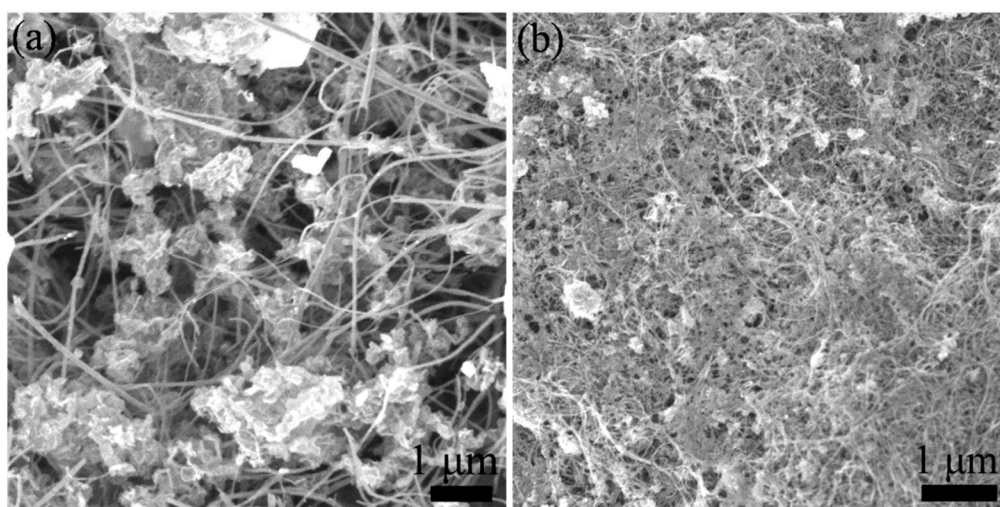
**Figure S1** (a) SEM and (b) TEM images of long- skeletal CNTs. This kind of CNTs has the length up to half a millimeter level and the diameter at 30-50 nm. More details of such CNTs and their treatments are included in reference S1.



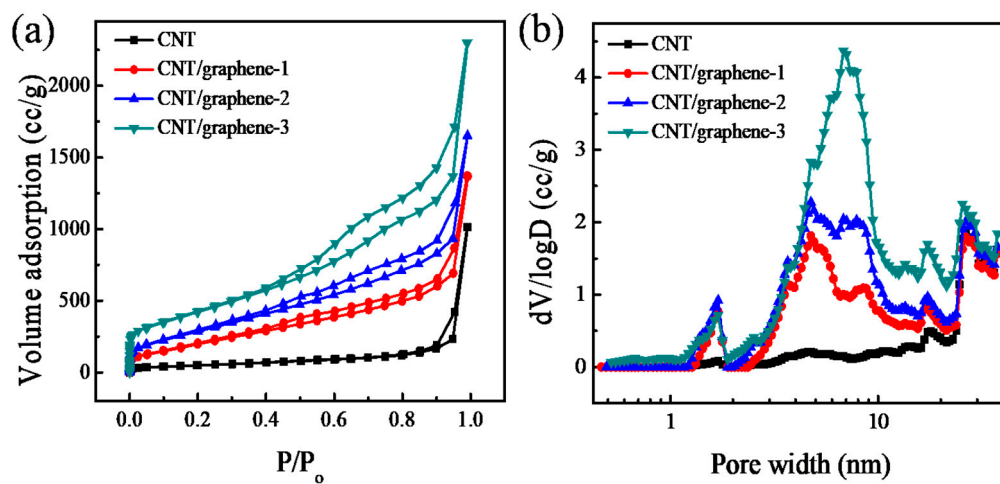
**Figure S2** (a) Low- and (b) high- magnification TEM images of short-capillary CNTs. This kind of CNTs has the length up to 100 micrometers and diameter of  $\sim 11$  nm. More details of such CNTs and their treatments are included in reference S2.



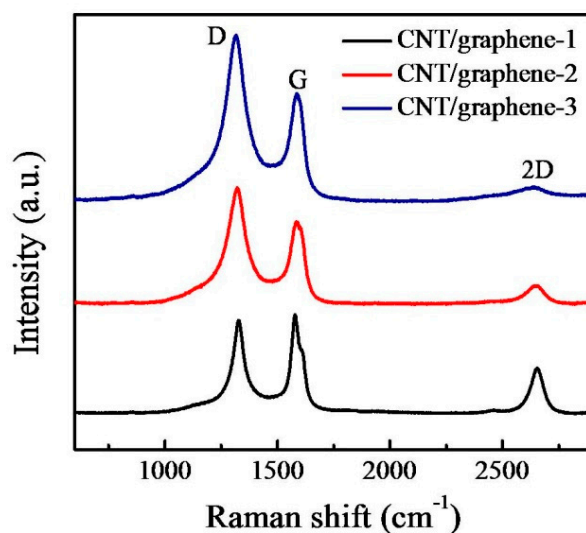
**Figure S3** (a) TEM image and (b) XRD pattern of  $\text{Mg(OH)}_2$  powder. It displays nanosheet morphology, with typical diffraction peaks.



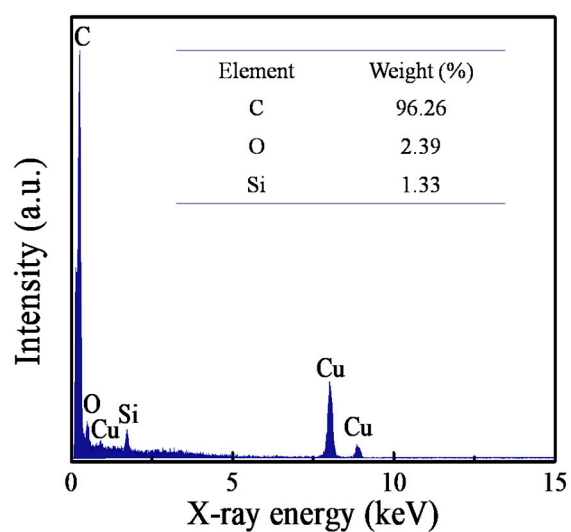
**Figure S4** (a) SEM image of long-skeletal CNT/graphene nanocomposite, showing a loose phase separation. (b) SEM image of short-capillary CNT/graphene nanocomposite, showing tight integration with partial graphene aggregates.



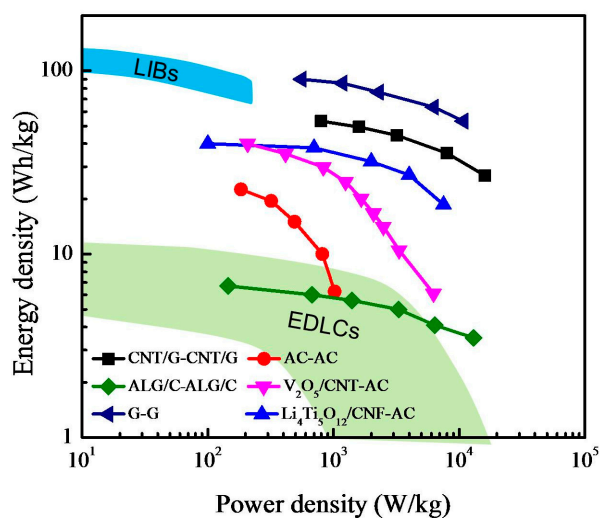
**Figure S5** (a)  $N_2$  sorption isotherms and (b) pore size distribution of the CNT and CNT/graphene composites with different compositions.



**Figure S6** Raman spectrum of the as-prepared CNT/graphene composites.



**Figure S7.** EDX spectrum of the skeletal-capillary CNT/graphene nanocomposite, showing the C and O lines, and Cu sample holder line.



**Figure S8** Comparison of Ragone plots of the ternary CNT/graphene supercapacitor with other devices, including symmetric supercapacitors made from ALG-C [S3], AC [S4] or graphene [S5], and asymmetric supercapacitors based on a  $V_2O_5/CNT$  [S4] or  $Li_4Ti_5O_{12}/CNF$  [S6] nanocomposite anode and AC cathode. All the data are based on the total mass of electrode materials.

**Table S1** The composition and N<sub>2</sub> sorption characterization of the nanocomposites.

Sample	Composition (CNT skeleton: CNT capillary: Mg(OH) <sub>2</sub> )	Composition* (CNT skeleton: CNT capillary: graphene, %)	Surface area (m <sup>2</sup> /g)	Pore volume (cm <sup>3</sup> /g)
CNTs	1:2:0	33:67:0	169.1	0.76
CNT/graphene-1	1:2:26	18:37:45	668.1	1.44
CNT/graphene-2	1:2:54	12:25:63	959.3	1.86
CNT/graphene-3	1:2:93	8:17:75	1453.7	2.72

\* 9.5 wt-% carbon yield based on the weight of Mg(OH)<sub>2</sub>.

## References

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- [S6] Naoi, K. 'Nanohybrid Capacitor': The Next Generation Electrochemical Capacitors. *Fuel Cells* 2010, 10, 825-833.