

## Supporting Information

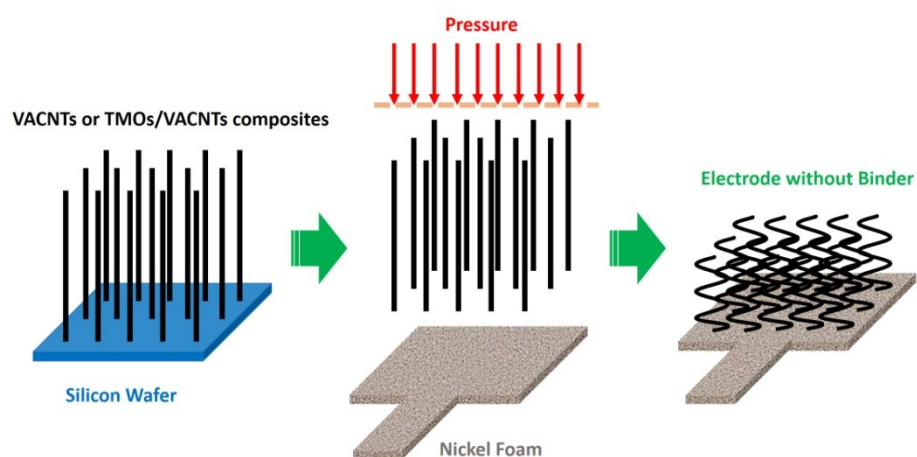
### **Fe<sub>2</sub>O<sub>3</sub>--Decorated Millimeter-Long Vertically Aligned Carbon Nanotube Arrays as Advanced Anode Materials for Asymmetric Supercapacitor with High Energy and Power Densities**

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Yan<sup>1</sup>, Junhe Yang<sup>1</sup>, Jianqiang Li<sup>2,\*</sup>

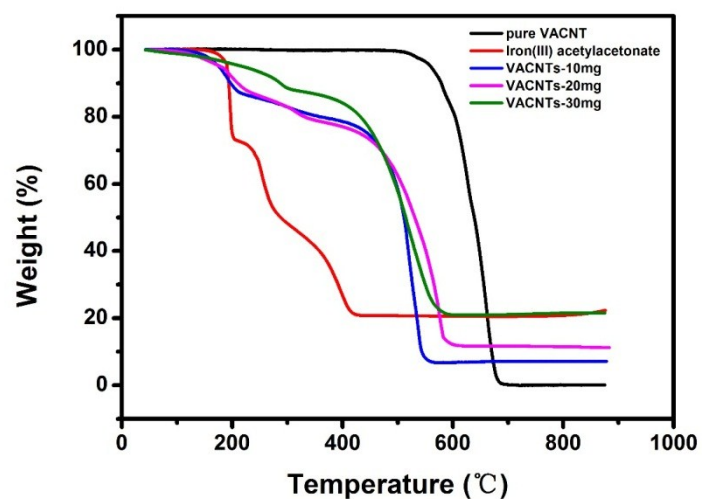
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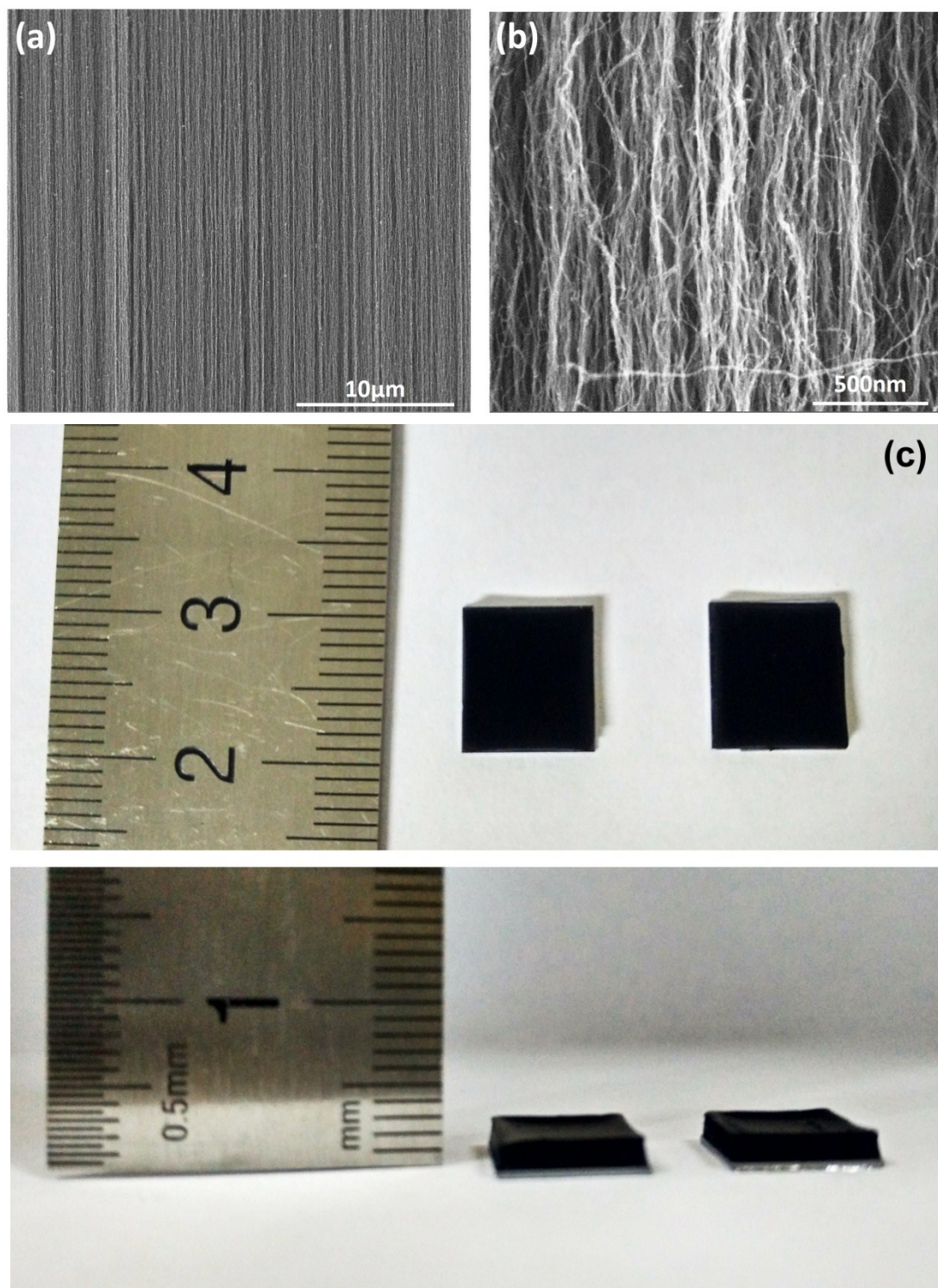
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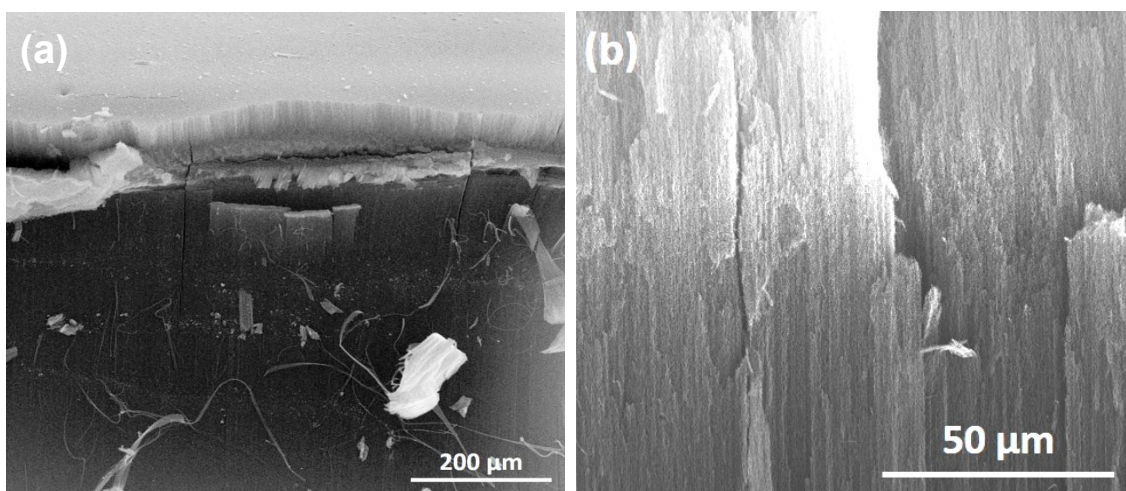
**Figure S1.** Schematic diagram of the electrode fabrication for electrochemical measurements.



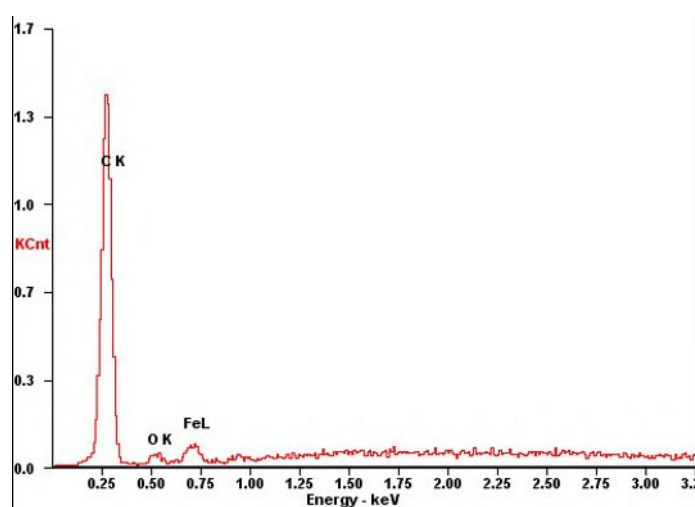
**Figure S2.** Thermogravimetric analysis of the pristine VACNT (black), the iron precursor (red), VACNTs with 10mg precursor (blue), VACNTs with 20mg precursor (pink), and VACNTs with 30mg precursor (green).



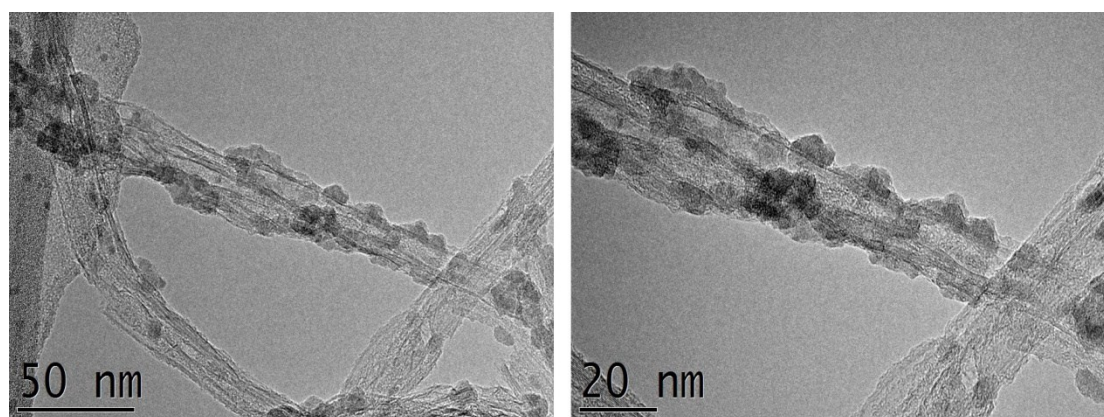
**Figure S3.** Cross-sectional SEM images (a, b) and digital photograph (c) of the pristine VACNTs. (Thickness of Si substrate: 0.5 mm.)



**Figure S4.** Cross-sectional SEM images of the VACNTs-26.06 composite: (a) low-magnification; (b) magnified image for the top surface layer.



**Figure S5.** Corresponding EDS spectrum for the selected section in Fig. 2e.



**Figure S6.** TEM images of the VACNTs-17.10 sample showing the decoration of  $\text{Fe}_2\text{O}_3$  particles on outer surface of CNTs.

**Table S1 .** Effect of impregnation time in  $\text{SCCO}_2$  on the weight gain of VACNTs.

| Impregnation time in $\text{SCCO}_2$ | 1h      | 3h      | 6h      | 9h      | 12h     |
|--------------------------------------|---------|---------|---------|---------|---------|
| Weight gain of VACNTs                | 0.57 mg | 1.28 mg | 2.37 mg | 2.56 mg | 2.78 mg |

**Table S2.** Effect of precursor mass on the weight gain of VACNTs after  $\text{SCCO}_2$  processing and the weight percentage of  $\text{Fe}_2\text{O}_3$  in the composite.

| Mass of Fe precursor | Weight gain of VACNTs after impregnation | Wt.% of $\text{Fe}_2\text{O}_3$ in the composite after annealing |
|----------------------|------------------------------------------|------------------------------------------------------------------|
| 10 mg                | 0.83 mg                                  | 5.41                                                             |
| 20 mg                | 1.60 mg                                  | 13.95                                                            |
| 30 mg                | 2.37 mg                                  | 17.10                                                            |
| 40 mg                | 3.17 mg                                  | 26.06                                                            |

**Table S3.** Parameters for capacitance calculation in Fig.4b

| Sample                  | Voltage drop (V) | Discharge time (s) |
|-------------------------|------------------|--------------------|
| VACNTs                  | 1.2              | 2.86               |
| $\text{Fe}_2\text{O}_3$ | 1.2              | 19.20              |

|              |     |       |
|--------------|-----|-------|
| VACNTs-5.41  | 1.2 | 20.34 |
| VACNTs-13.95 | 1.2 | 28.67 |
| VACNTs-17.10 | 1.2 | 37.20 |
| VACNTs-26.06 | 1.2 | 30.15 |

**Table S4.** Parameters for capacitance calculation in Fig.5e

| Current density (A g <sup>-1</sup> ) | Voltage drop (V) | Discharge time (s) |
|--------------------------------------|------------------|--------------------|
| 2                                    | 0.6              | 181.76             |
| 4                                    | 0.6              | 178.72             |
| 6                                    | 0.6              | 174.69             |
| 8                                    | 0.6              | 171.76             |
| 12                                   | 0.6              | 170.52             |
| 16                                   | 0.6              | 169.6              |
| 20                                   | 0.6              | 168.6              |
| 24                                   | 0.6              | 167.98             |

**Table S5.** Parameters for capacitance calculation in Fig.6c

| Current density (A g <sup>-1</sup> ) | Discharge time (s) | IR Drop (V) |
|--------------------------------------|--------------------|-------------|
| 2                                    | 64                 | 0.12        |
| 4                                    | 28                 | 0.2         |
| 6                                    | 17                 | 0.22        |
| 8                                    | 12.5               | 0.22        |
| 12                                   | 8                  | 0.27        |
| 16                                   | 5.7                | 0.31        |
| 20                                   | 4                  | 0.45        |
| 24                                   | 2.6                | 0.7         |

**Table S6.** Volume capacitance, volume power density/energy density of the asymmetric supercapacitor

| Current density<br>(mA/cm <sup>3</sup> ) | Volume specific<br>capacitance<br>(F/cm <sup>3</sup> ) | Volume energy<br>density (mWh/cm <sup>3</sup> ) | Volume power<br>density (W/cm <sup>3</sup> ) |
|------------------------------------------|--------------------------------------------------------|-------------------------------------------------|----------------------------------------------|
| 90                                       | 13.7                                                   | 6.2                                             | 0.1                                          |
| 180                                      | 12.6                                                   | 5.7                                             | 0.2                                          |
| 270                                      | 11.6                                                   | 5.2                                             | 0.3                                          |
| 360                                      | 11.4                                                   | 5.1                                             | 0.4                                          |
| 540                                      | 11.3                                                   | 5.1                                             | 0.6                                          |
| 720                                      | 11.0                                                   | 5.0                                             | 0.9                                          |
| 900                                      | 10.7                                                   | 4.8                                             | 1.2                                          |
| 1080                                     | 10.2                                                   | 4.6                                             | 1.8                                          |

\* In the calculation, whole volume of the asymmetric supercapacitor is used, including the active materials, the current collector, the separator and the electrolyte.