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Electronic Supplementary Information for

Atomic Layer Deposition of Nickel Carbide for Supercapacitors and Electrocatalytic Hydrogen Evolution

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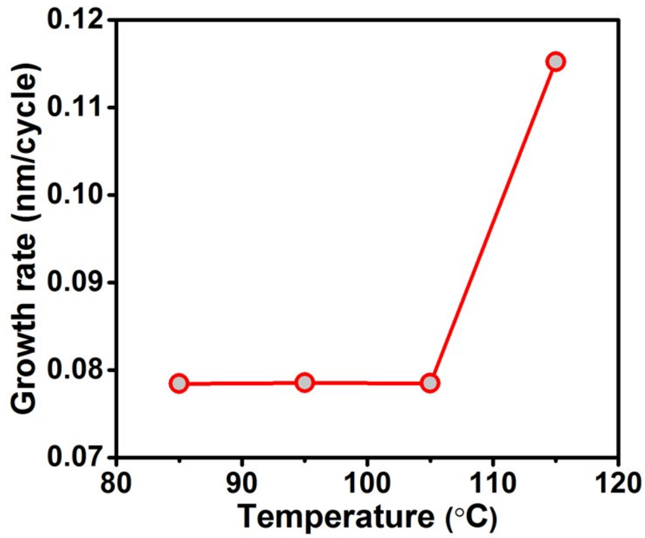

Figure S1. ALD growth rate of Ni_3C as a function of deposition temperature.

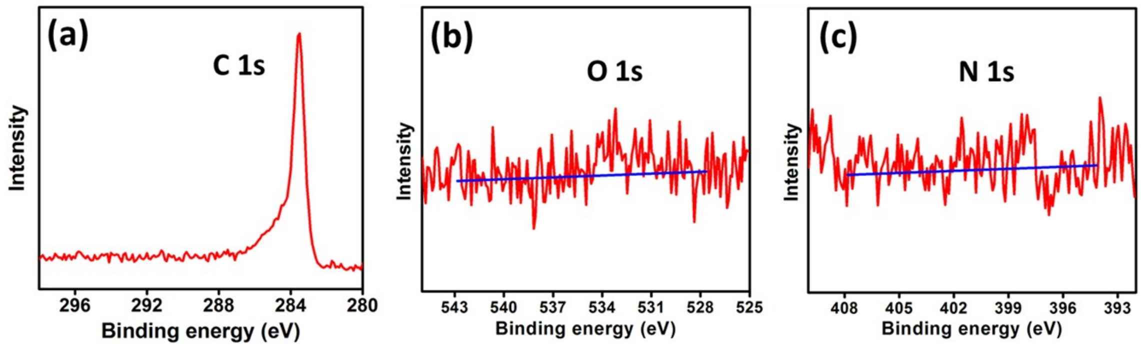

Figure S2. High-resolution XPS spectra of (a) C 1s, (b) O 1s, and (c) N 1s for the ALD Ni_3C film.

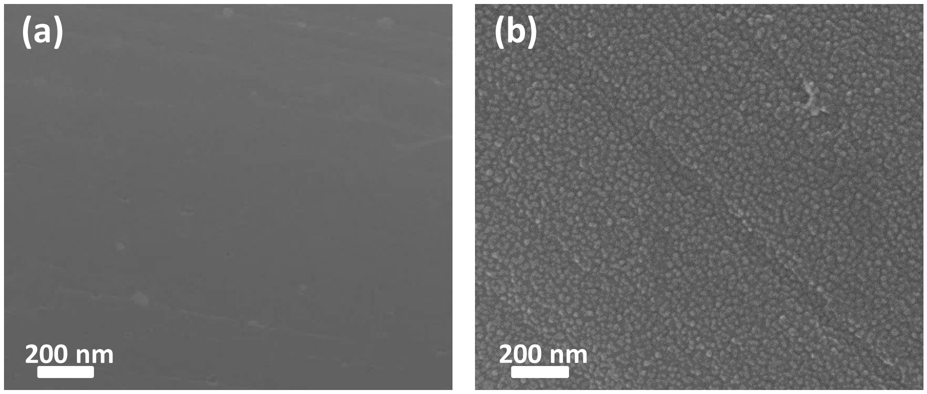

Figure S3. High-resolution SEM images of (a) pristine CC and (b) ALD Ni_3C coated CC.

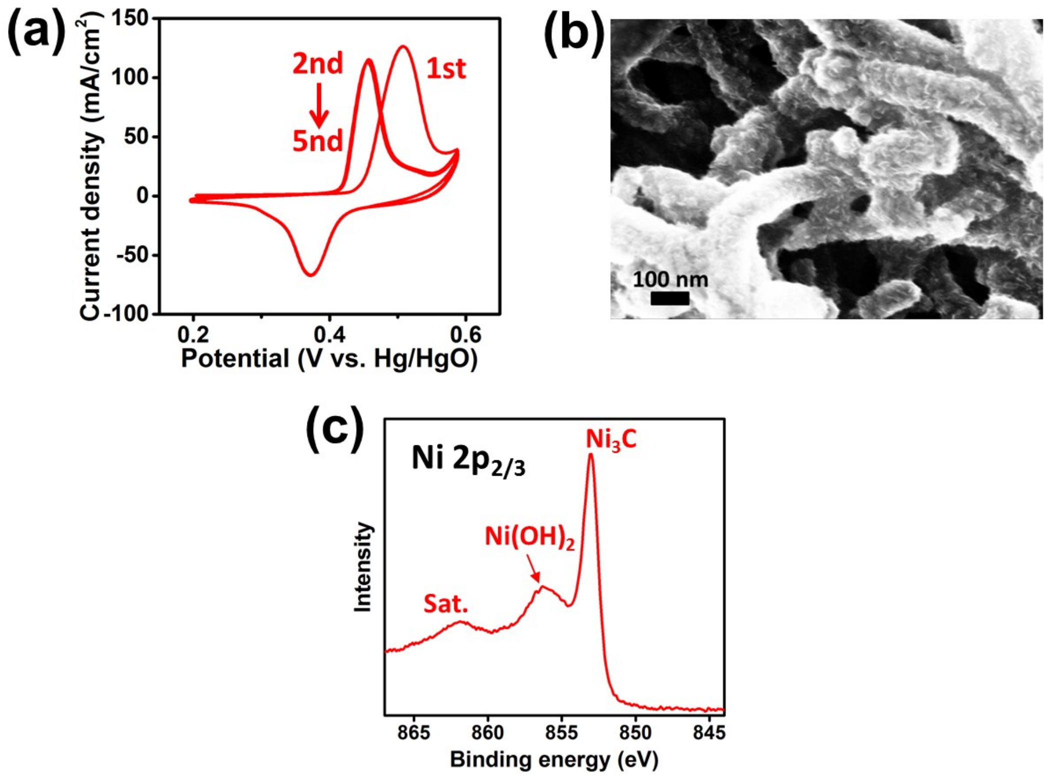

Figure S4. (a) CV curves for the initial 6 cycles of the ALD Ni_3C/CNT electrode (scan rate 30 mV/s). The first-cycle curve is apparently different from the following cycles, which implies possible irreversible oxidation of the surface Ni_3C ($\frac{1}{3}$ Ni_3C + $3OH^- \rightarrow$ NiOOH + H_2O + $\frac{1}{3}$C+ $3e^-$) during the first anodic scan (*J. Solid State Electrochem*. 2016, **20**, 775). The subsequently CV curves show a reversible redox pair centered at 0.41 V, which corresponds to the redox reaction between $Ni(OH)_2$/NiOOH ($Ni(OH)_2$ + $OH^- \leftrightarrow$ NiOOH + H_2O + e^-). (*Electrochim. Acta,* 2016, **204**, 160−168; *J. Mater. Chem. A*, 2013, **1**, 7880−7884; *Adv. Funct. Mater.*, 2012, **22**, 1272−1278) (b) SEM image of the Ni_3C/CNT after CV cycling for 5000 cycles. On the Ni_3C/CNT surface formed some nanoflakes, which were representative for $Ni(OH)_2$ (*Adv. Funct. Mater.*, 2012, **22**, 1272−1278; *J. Mater. Chem. A*, 2015, **3**, 19545−19555). (c) XPS Ni $2p_{3/2}$ spectrum taken after the 5000-cycle CV, in which a peak corresponding to $Ni(OH)_2$ was observed (*Adv. Funct. Mater.*, 2015, **25**, 7530–7538), suggesting the formation of the $Ni(OH)_2$ on the Ni_3C surface.

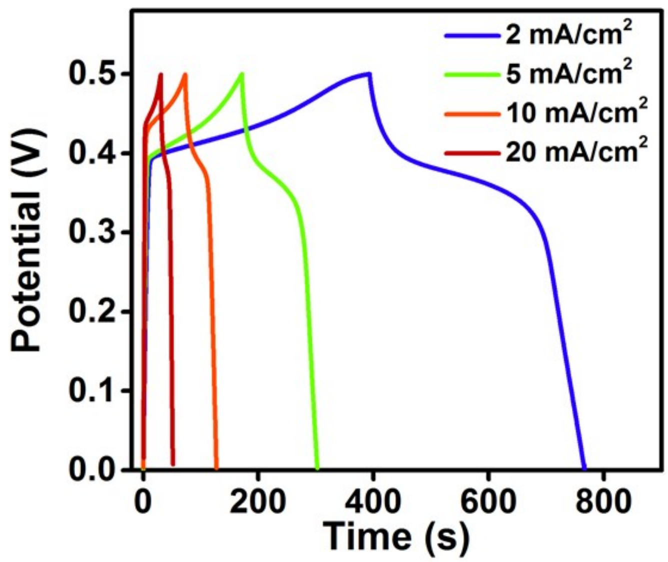

Figure S5. Charge-discharge curves of the ALD Ni_3C/CNT electrode at various current densities.

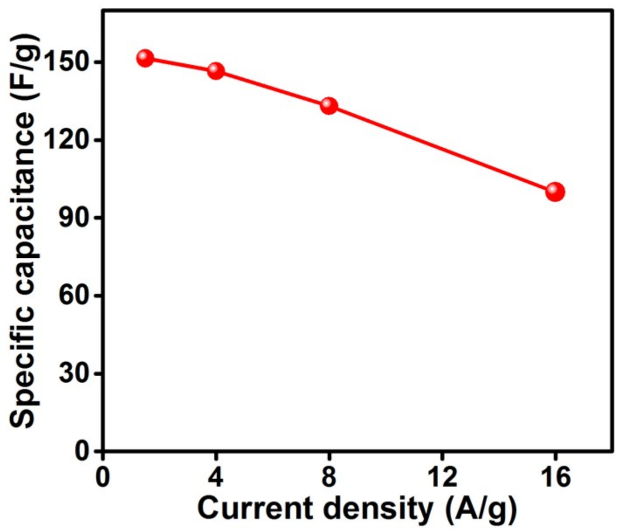

Figure S6. Specific capacitance of the Ni_3C/CNT//AC supercapacitor with respect to discharge current density.

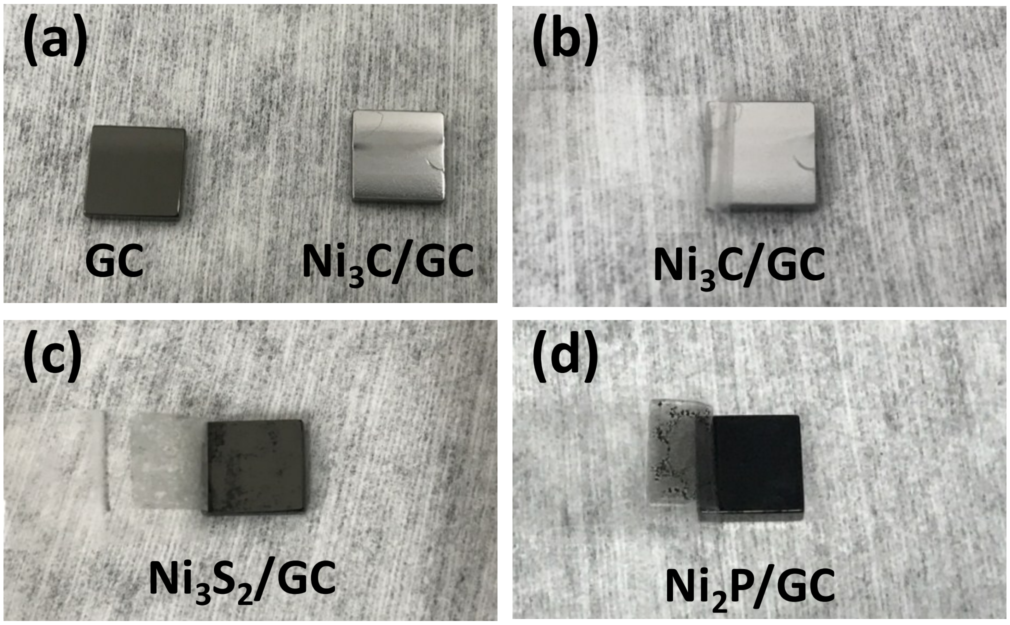

Figure S7. (a) Photograph of glassy carbon (GC) and ALD Ni_3C coated GC (Ni_3C/GC). Photographs of tape tests for (b) Ni_3C/GC (passed), (c) Ni_3S_2/GC (failed), and (d) Ni_2P/GC (failed). Ni_3S_2/GC was synthesized by electrodeposition of Ni_3S_2 on GC (Xing, *et al.*, *J. Power Sources* **2014**, *245*, 463–467), and Ni_3S_2/GC was synthesized by phosphorization of $Ni(OH)_2$ on GC (Zhou, *et al.*, *Adv. Funct. Mater.* **2015**, *25*, 7530–7538).

Table S1. Comparison of the supercapacitor performance for the ALD Ni_3C/CNT and various other reported Ni-based materials.

Active materials	Specific capacitance	Capacitance retention upon cycling	Reference
Ni_3C/CNT	**1850 F/g (at 4 A/g)**	**98.5% (after 5000 cycles at 40 A/g)**	**This work**
$Ni(OH)_2$/Ni foam	2384 F/g (at 1 A/g)	75% (after 1000 cycles at 5 A/g)	*Nano Energy* **2015**, *11*, 154
Mg-$Ni(OH)_2$	1931 F/g (at 0.5A/g)	95% (after 10000 cycle at 10 A/g)	*ACS Energy Lett.,* **2016**, *1*, 814
Ni_3S_2/3D-rGO	1886 F/g (at 1 A/g)	91% (after 30000 cycle at 2 A/g)	*ACS Energy Lett.,* **2017**, *2,* 759
$Ni(OH)_2$/g-C_3N_4	1768 F/g (at 7 A/g)	84 % (after 8000 cycle at 51 A/g)	*ACS Appl. Mater. Interfaces* **2017**, *9*, 17890
Ni-P	1597 F/g (at 0.5 A/g)	71.4% (after 1000 cycles at 2 A/g)	*J. Power Sources* **2015**, *274*, 1107
$NiMoO_4$ nanowires	1587 F/g (at 6 A/g)	76.9% (after 4000 cycles at 18 A/g)	*Nano Energy* **2014**, *8*, 174
$Ni(OH)_2$/graphite	1560 F/g (at 0.5 A/g)	65% (after 1000 cycles at 10 A/g)	*ACS Nano* **2013**, *7*, 6237
NiO/GO foam	1225 F/g (at 2 A/g)	94% (after 2000 cycles at 12 A/g)	*J. Mater. Chem. A* **2014**, *2*, 3223
$NiCo_2O_4$ hollow spheres	1141 F/g (at 1 A/g)	94.7% (after 4000 cycles at 5 A/g)	*Angew. Chem. Int. Ed* **2014**, *53*, 1488
$NiCo_2S_4$ hollow spheres	1036 F/g (at 1 A/g)	87% (after 2000 cycles at 5 A/g)	*Nat. Commun.* **2015**, *6*, 6694
Ni_3N/CC	990 F/g (at 3.5 A/g)	50% (after 2000 cycles at 100 mV/s)	*J. Mater. Chem. A* **2016**, *24*, 9844
NiS/rGO	905 F/g (at 0.5 A/g)	90% (after 2000 cycles at 4 A/g)	*Nano Energy* **2014**, *5*, 74
$Ni_{0.85}Se/MoSe_2$	774 F/g (at 1 A/g)	95% (after 1000 cycles at 3 A/g)	*ACS Appl. Mater. Interfaces* **2017**, *9*, 17067
Ni-B/carbon fiber	733 F/g (at 5 mV/s)	72% (after 1000 cycles at 10 mV/s)	*J. Mater. Sci,* **2015**, *50*, 4622
CoNiAl-LDH/AC	501 F/g (at 10 A/g)	91% (after 1000 cycles at 6 A/g)	*Nanoscale* **2014**, *6*, 3097

Table S2. Comparison of the HER performance of the ALD Ni_3C/CNT with various other nonprecious catalysts in alkaline solution.

Active materials	Overpotential ($j = -10$ mA/cm^2)	Tafel slope (mV/dec)	Long-term stability (current retention under constant bias)	Reference
Ni_3C/CNT	**−132 mV**	**49**	**93% after 48 h (initial *j* = −10 mA/cm^2)**	**This work**
NiO/Ni-CNT	−80 mV	82	95% after 2 h (initial *j* = −5 mA/cm^2)	*Nat. Commun.* **2014**, *5*, 4695
$Ni_{1-x}Fe_x$/Nanocarbon	−184 mV	100	91% after 0.3 h (initial *j* = −20 mA/cm^2)	*ACS Catal.* **2016**, *6*, 580
Ni/N-Graphene	−205 mV	160	90.5% after 10 h (initial *j* = −10 mA/cm^2)	*Adv. Mater.* **2017**, *26*, 1605957
$Co_{0.85}Se$/NiFe-LDH	−265 mV	160	86% after 10 h (initial *j* = −10 mA/cm^2)	*Energy Environ. Sci.* **2016**, *9*, 478
Ni/NiS	−230 mV	115	84% after 12 h (initial *j* = −10 mA/cm^2)	*Adv. Funct. Mater.* **2016**, *26*, 3314
Co-C-N complex	−178 mV	102	80% after 40 h (initial *j* = −30 mA/cm^2)	*J. Am. Chem. Soc.* **2015**, *137*, 15070
Mo_2C	−190 mV	54	80% after 48 h (initial *j* = −17 mA/cm^2)	*Angew. Chem.* **2012**, *124*, 12875
Ni_5P_4	−150 mV	59	50% after 20 h (initial *j* = −10 mA/cm^2)	*Angew. Chem.* **2015**, *54*, 12361
MoC_x	−151 mV	75	27% after 11 h (initial *j* = −15 mA/cm^2)	*Nat. Commun.* **2015**, *6*, 6512
MoP	−169 mV	70	15% after 10 h (initial *j* = −10 mA/cm^2)	*Nano Energy* **2017**, *32*, 511
MoP	−125 mV	48	60% after 40 h (initial *j* = −15 mA/cm^2)	*Energy Environ. Sci.,* **2014**, *7*, 2624
$MoSe_2$@$Ni_{0.85}Se$	−117 mV	66	77% after 20 h (initial *j* = −20 mA/cm^2)	*Electrochim. Acta* **2017**, *246*,712