Electronic Supplementary Material (ESI) for Nanoscale.

Electronic Supplementary Information (ESI) for:

Amorphous nickel sulfides nanoparticles anchored on the N-doped graphene nanotubes with superior properties for high-performance supercapacitor and efficient oxygen evolution

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Calculations:

(1) The specific capacitances (C_g) of the N-GNTs@NSNs on GS electrode calculated from GCD curves are obtained according to the following equation:

$$C_g = \frac{I\Delta t}{m\Delta V}$$

where I is the discharge current, Δt is the discharge time in GV test, m is the mass loading of the electrode materials, s is the area of the electrode materials, and ΔV is the voltage window.

(2) The specific capacitance (C_{device}) of the N-GNTs@NSNs on GS// AC on NF asymmetric supercapacitor (ASC) device can be obtained in accordance with the following equation:

$$C_{\mathrm{de}vice} = \frac{I\Delta t}{M\Delta V}$$

Herein, I is the discharge current, Δt is the discharge time in GCD test, M is the mass of the device, and ΔV is the voltage window of the device.

(3) Methods to calculate the energy and power density of the ASC device:

$$\mathrm{E} = \frac{1}{7.2}\mathrm{C}_{\mathrm{device}}\Delta\mathrm{V}^2;\ \mathrm{P} = \frac{\mathrm{E}}{\mathrm{t}}$$

Here, C_{device} is the specific capacitance of the device, ΔV is the potential window, and t is the discharge time.

(4) The measured potentials are referred to RHE using the following equation:

$$E(\mathrm{RHE}) = E\ (\mathrm{SCE}) + 0.059\mathrm{PH} + 0.241$$

(5)The electrochemical double-layer capacitance (Cdl) is determined from the CV curves measured in a potential range without redox process by following equation:

$$Cdl = \frac{I}{v}$$

where I is the charging current (mA cm^{-2}), and v is the scan rate (mV s^{-1}).

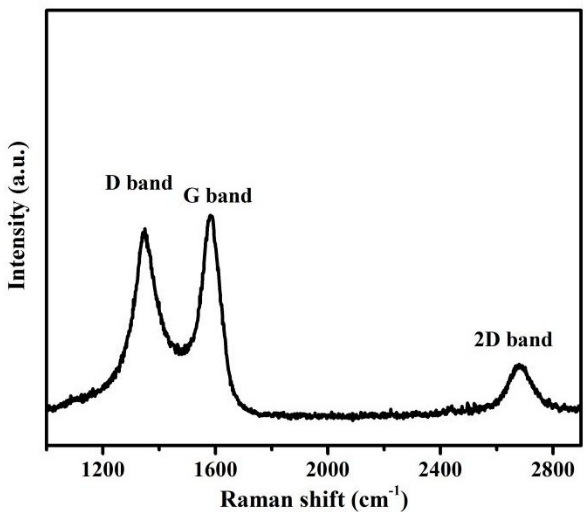

Fig. S1 Raman spectrum of the pure N-GNTs.

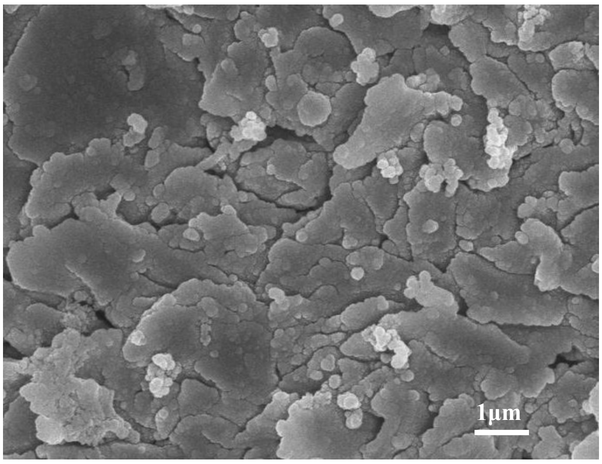

Fig. S2 Low-magnification SEM images of nickel sulfides without N-GNTs.

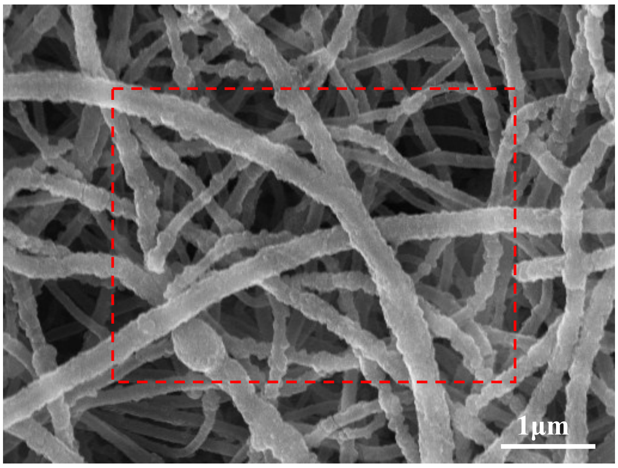

Fig. S3 SEM images of N-GNTs@NSNs.

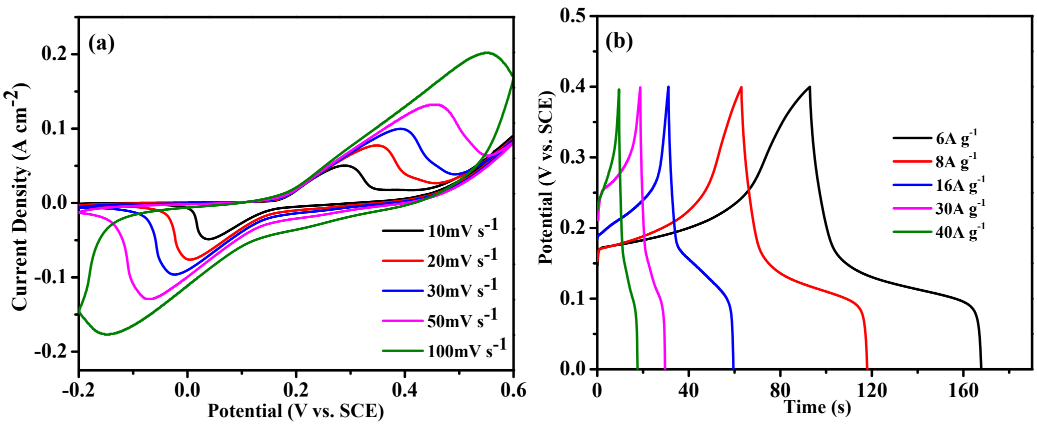

Fig. S4 (a) CV and (b) GCD curves of the NSNs electrode in 3 M KOH aqueous electrolyte.

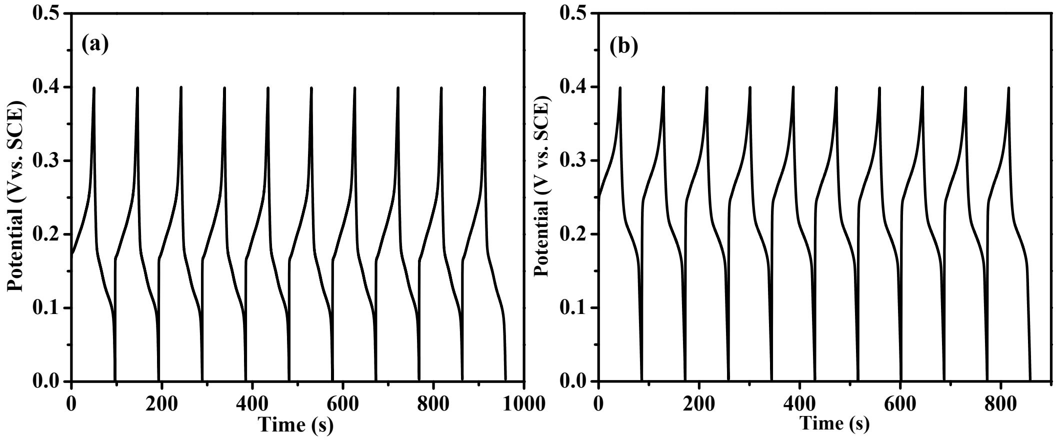

Fig. S5 Galvanostatic results of (a) the first ten cycles and (b) the last ten cycles of the N-GNTs@NSNs hybrid electrodes.

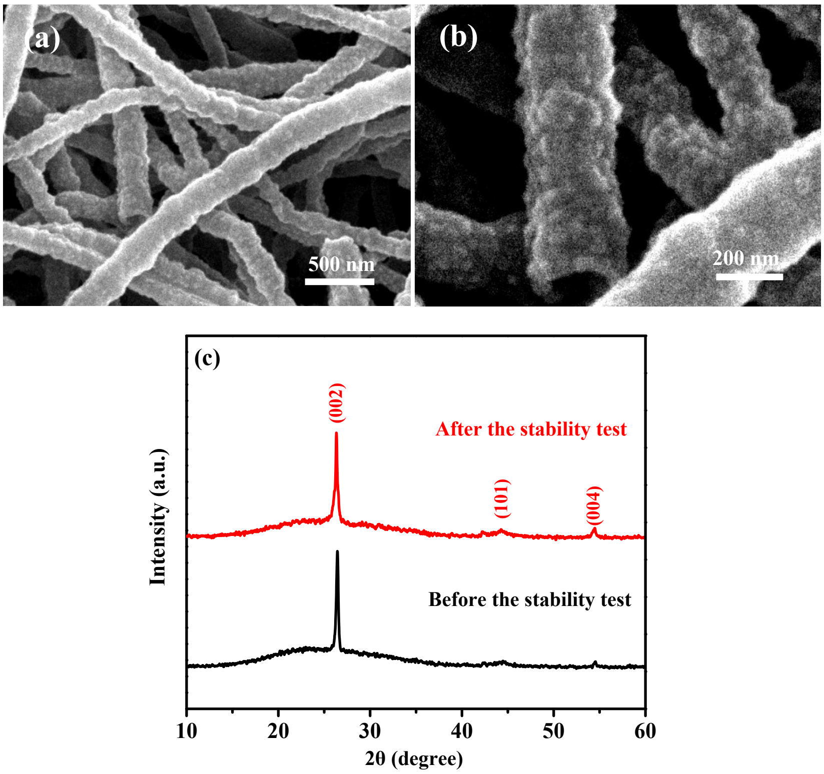

Fig. S6 SEM images of the N-GNTs@NSNs after 12000 cycles.

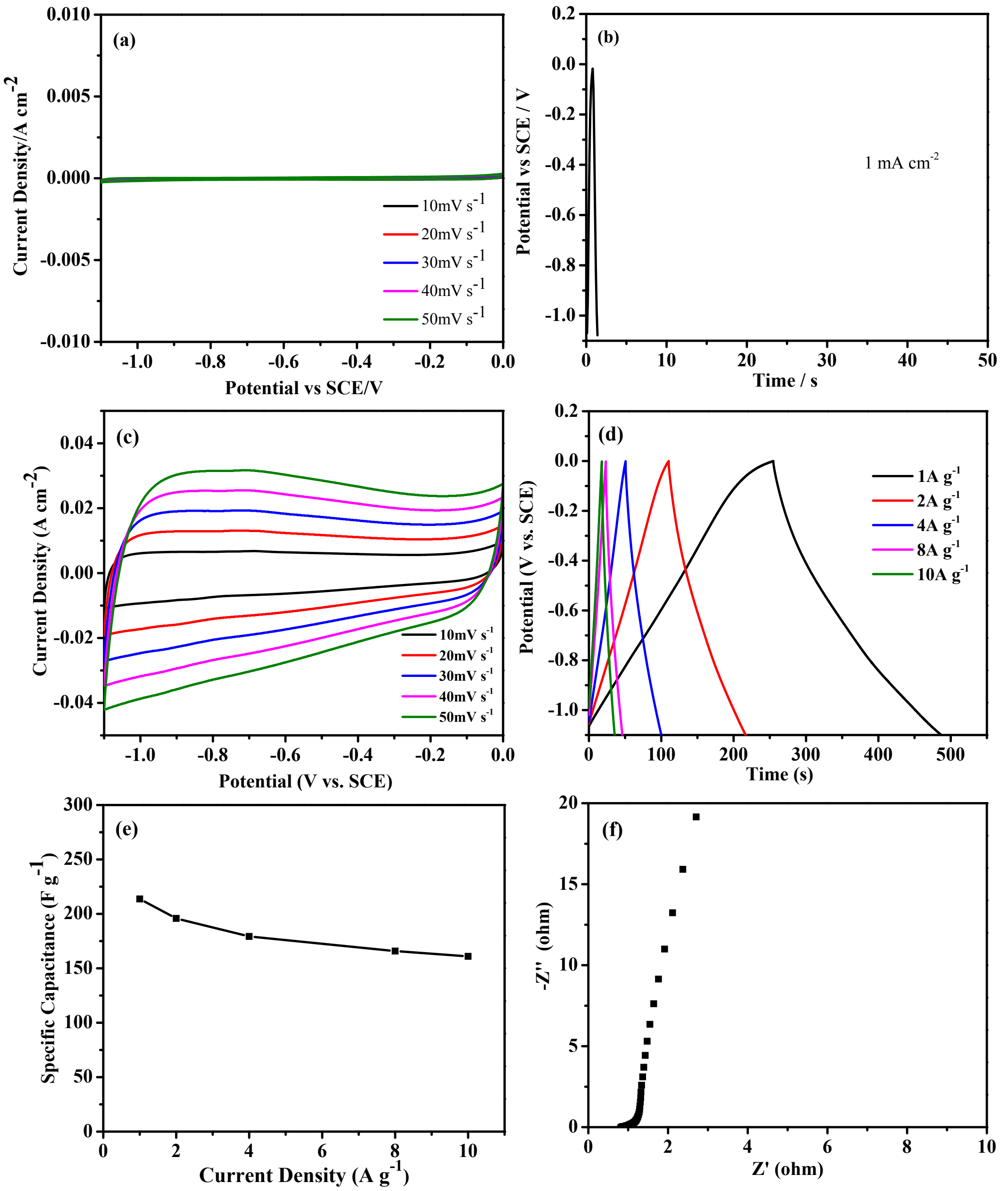

Fig. S7 (a) CV curves of nickel foam at different scan rates. (b) GCD curves of nickel foam at 1 mA cm^{-2}. (c) CV curves of AC at different scan rates. (d) GCD curves of AC at different current densities. (e) Specific capacity at different current densities from 1 to 10 A g^{-1}. (f) Nyquist plot of the AC electrode.

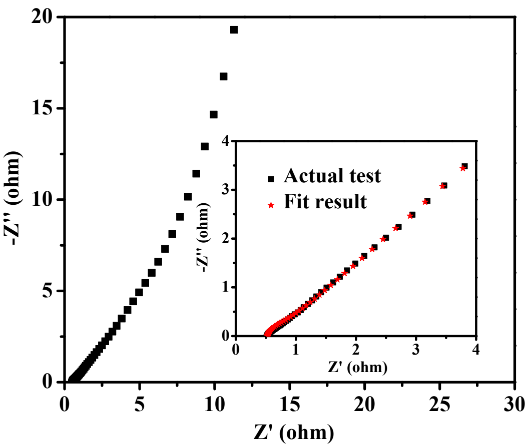

Fig. S8 Nyquist plots and an equivalent circuit diagram of the ASC device.

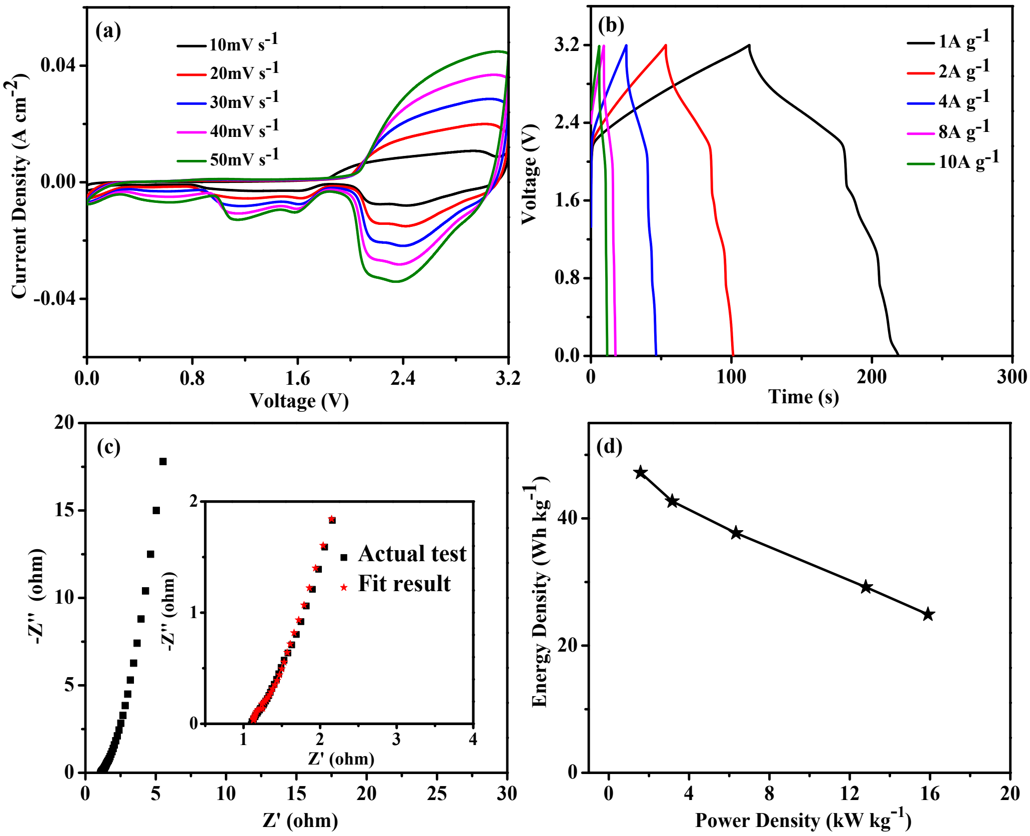

Fig. S9 (a) CV curves at various scan rates of device after series; (b) GCD curves at different current densities of 1-10 A g^{-1}; (c) Nyquist plot and (d) the Ragone plot.

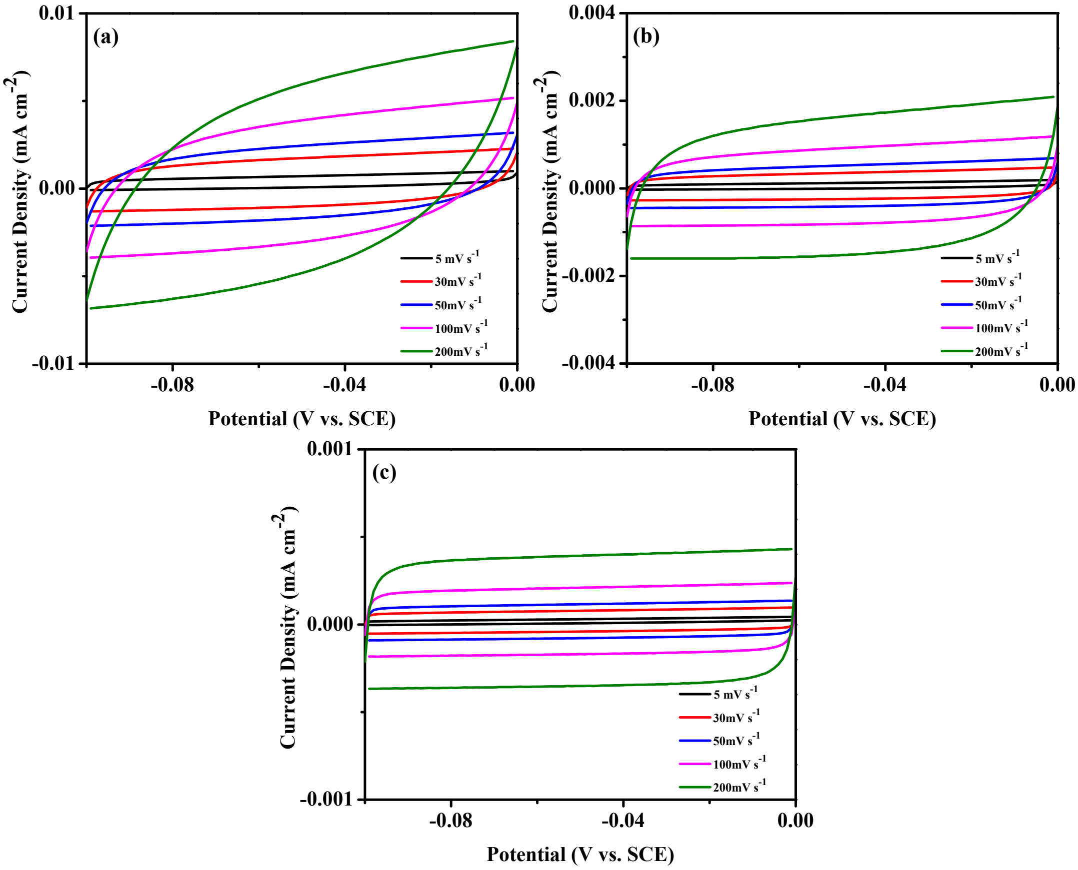

Fig. S10. CV curves of the (a) N-GNTs@NSNs, (b) N-GNTs and (c) NSNs electrode.

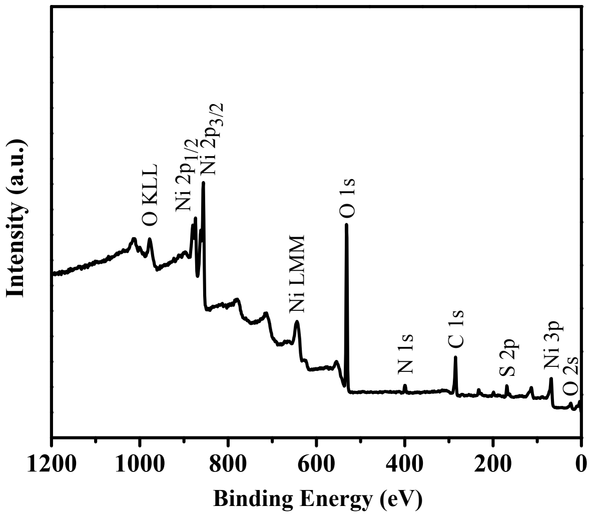

Fig. S11. XPS survey spectrum of the N-GNTs@NSNs after the stability test.

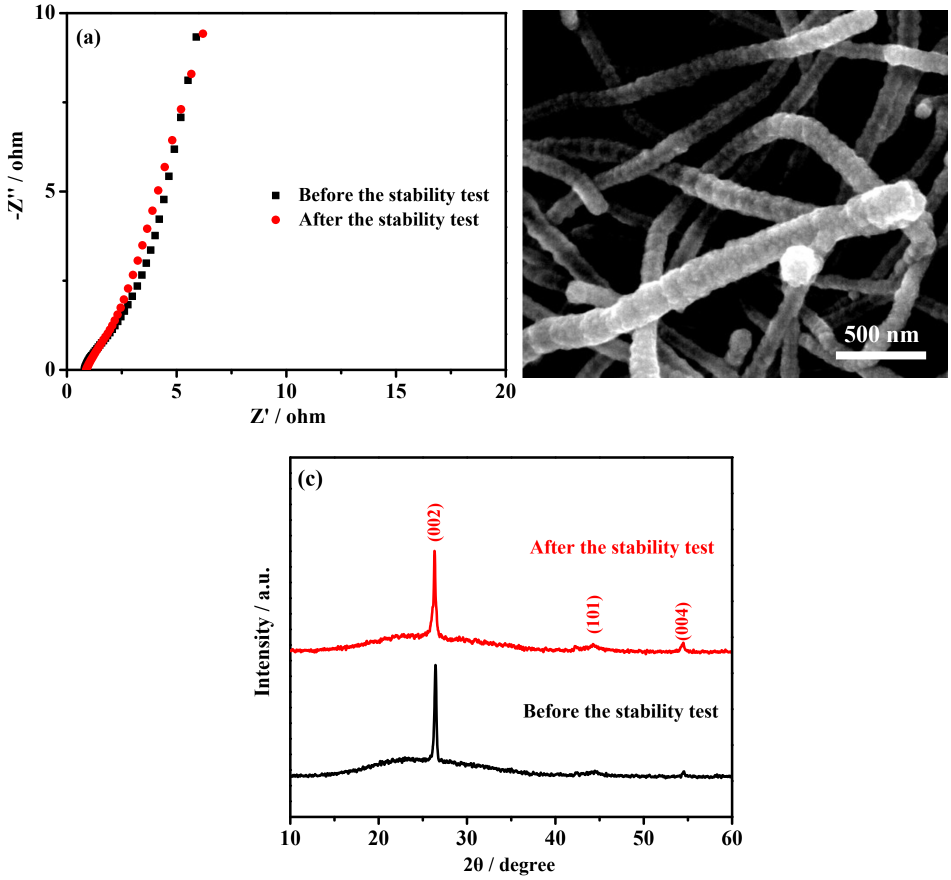

Fig. S12. (a) The Nyquist plots, (b) SEM image and (c) XRD pattern of the N-GNTs@NSNs after the stability test.

Tab. S1. Electrochemical performances comparison of the as-prepared N-GNTs@NSNs with the other Ni-based compound fabricated by different methods.

Material	Fabrication method	current collector	electrolyte	Specific capacitance	Rate performance	Reference
NiS cubes	Solvothermal and calcination	Ni foam	2M KOH	874.5F g^{-1}(1 A g^{-1})	454.5 F g^{-1}(20 A g^{-1})	S1
$NiCo_2S_4/Co_9S_8$	Hydrothermal	Ni foam	6M KOH	749F g^{-1} (4 A g^{-1})	620F g^{-1} (15 A g^{-1})	S2
Co_3O_4/Ni-based MOFs	Hydrothermal	Carbon cloth	3M KOH	209 mAh g^{-1} (2 A g^{-1})	58 mAh g^{-1} (10 A g^{-1})	S3
(Ni,Co)Se_2/NiCo-LDH	Calcination and electrodeposition	Carbon substrate	3M KOH	170 mAh g^{-1} (2 A g^{-1})	120.7 mAh g^{-1} (20 A g^{-1})	S4
Ni_3S_2@β-NiS Core–Shell	Solvothermal	Ni foam	2M KOH	1158F g^{-1} (2 A g^{-1})	670F g^{-1} (50 A g^{-1})	S5
GO@NiCo-LDH	Hydrothermal	Powder	6M KOH	1489F g^{-1} (1 A g^{-1})	1300F g^{-1} (20 A g^{-1})	S6
$NiCo_2O_4$@rGO	Hydrothermal	Ni foam	6M KOH	1125F g^{-1} (1 .5A g^{-1})	922F g^{-1} (6A g^{-1})	S7
NiCo-LDH	Hydrothermal	Graphene film	6M KOH	227mAh g^{-1} (0.5 A g^{-1})	175mAh g^{-1} (20 A g^{-1})	S8
$NiCo_2S_4$ NNSs	Hydrothermal,	Ni foam	4M KOH	1667F g^{-1} (1 A g^{-1})	1427F g^{-1} (25 A g^{-1})	S9
Ag-rGO/$Ni(OH)_2$	Hydrothermal,	Ni foam	5M KOH	1220F g^{-1} (1 A g^{-1})	901F g^{-1} (5 A g^{-1})	S10
N-GNTs@NSNs	**Electrodeposition**	**Graphite substrates**	**3M KOH**	**240 mAh g^{-1} (2160F g^{-}) at 6 A g^{-1}**	**183 mAh g^{-1} (1650F g^{-1})) at 40 A g^{-1}**	**This work**

Tab. S2. Cycling stability comparison of the N-GNTs@NSNs with the other Ni-based compound.

Material	current collector	electrolyte	Current density	Cyclic performance	Reference
NiS cubes	Ni foam	2M KOH	4 A g^{-1}	90.2%(after 3000 cycles)	S1
$NiCo_2O_4$@rGO	Ni foam	2M KOH	10mA cm^{-2}	90%(after 2000 cycles)	S7
Ag-rGO/$Ni(OH)_2$	Ni foam	5M KOH	2 A g^{-1}	92.6%(after 2000 cycles)	S10
NiMoO4@Ni-Co-S	Ni foam	2M KOH	20mA cm^{-2}	91.7%(after 6000 cycles)	S11
NiAl-LDH	Ni foam	6M KOH	20A g^{-1}	93.75%(after 10000 cycles)	S12
Ni-Co-P NNSs	Ni foam	3M KOH	2A g^{-1}	93%(after 8000 cycles)	S13
NiS/NHCS	Ni foam	2M KOH	5 A g^{-1}	76%(after4000 cycles)	S14
Mn-Co-LDH@$Ni(OH)_2$	Ni foam	3M KOH	20 A g^{-1}	90.9%(after 5000 cycles)	S15
$Ni_2P_2O_7$	Ni foam	1M KOH	50mA cm^{-2}	87%(after 5000 cycles)	S16
$NiCo_2O_4$	Ni foam	6M KOH	10 A g^{-1}	76.92%(after 10000 cycles)	S17
N-GNTs@NSNs	**Graphite substrates**	**3M KOH**	**16 A g^{-1}**	**95.8%(after 12000 cycles)**	**This work**

Tab. S3. Cycling performance comparison of our device with the previously reported ASC devices

ASC devices	Cell voltage (V)	Cycle performance	Reference
NiO NSs@CNTs@CuO NWAs//AC	1.55	83.6% retention after 4000 cycles	S18
Ni/NiO//CNTs-COOH	1.8	92.8% retention after 10000 cycles	S19
Ni-Co-S-W /NF//AC/NF	1.6	91.7% retention after 6000 cycles	S20
ZNCO@$Ni(OH)_2$ NWAs/CNTF//VN@C NWAs/CNTS	1.6	90.3% retention after 3000 cycles	S21
$Ni(OH)_2$/CNs//AC	1.6	93% retention after 10000 cycles	S22
Ni-Mo-S NS//Ni-Fe-S NS	1.6	95.86% retention after 10000 cycles	S23
C@NiMn-OH-Ni_3S_2/Ni foam//AC	1.7	94.6% retention after 10000 cycles	S24
Ni_3S_2@Ni‖AC	1.8	62.5% retention after 10000 cycles	S25
CC/H-Ni@Al-Co-S//graphene/CNT	1.8	90.6% retention after 10000 cycles	S26
ZCS/$Ni(OH)_2$//ZCS/$Ni(OH)_2$	1.3	78% retention after 10000 cycles	S27
N-GNTs@NSNs//AC	**1.6**	**96.6% retention after 12000 cycles**	**In this work**

Tab. S4. Comparison of OER performances of Ni-Co compound based materials.

Catalysts	Powder/on substrates	Electrolyte	Overpotential @10mAcm^{-2} (mV)	Tafel slope (mV dec^{-1})	Reference
$NiCo_2O_4$	Carbon Cloth	1M KOH	340	72	S28
$Co_{0.85}Se$	carbon fabric collector	1M KOH	320	85	S29
NiCo-LDH	Graphene film	1M KOH	289	93.5	S8
CoO	Carbon Cloth	1M KOH	320	80	S30
CoCr@NGT	Glassy carbon	1M KOH	330	95	S31
$Co(CO_3)_{0.5}(OH)$	Ni foam	1M $KHCO_3$	332	126	S32
β-$Co(OH)_2$	Co Plate	1M KOH	332	68.3	S33
$Fe(TCNQ)_2$	Fe film	1M KOH	340	94	S34
Co_3O_4	Glassy carbon	1M KOH	307	76	S35
$Ni(OH)_2$-Ag-RGO	Glassy carbon	1M KOH	292	58	S36
N-GNT@NSNs	**Graphite substrates**	**1M KOH**	**284**	**60.7**	**This work**

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