Electronic Supplementary Material (ESI) for Nanoscale.


# Supporting Information

## Highly Stable, Stretchable, and Versatile Electrodes by Coupling of NiCoS Nanosheets with Metallic Networks for Flexible Electronics

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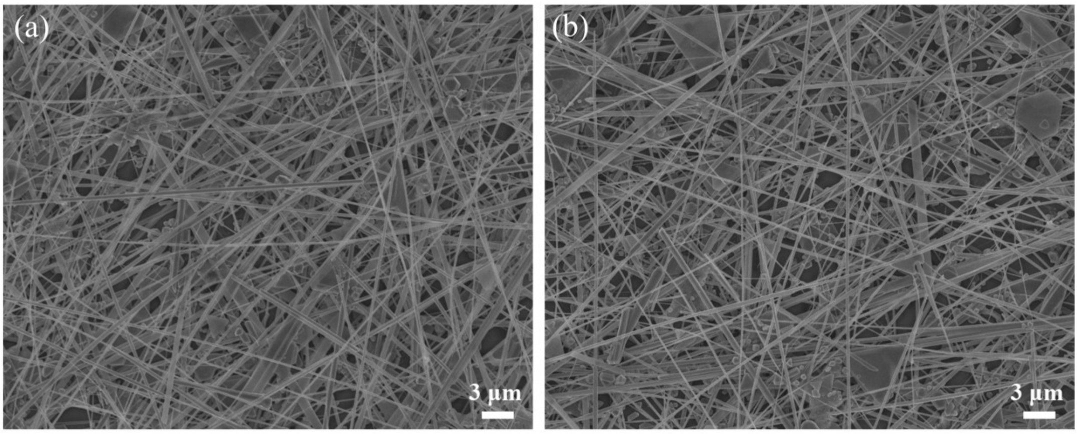


Figure S1 SEM images of the Ag NWs on a glass substrate (a) before and (b) after heat treatment at 300 °C for 30 min

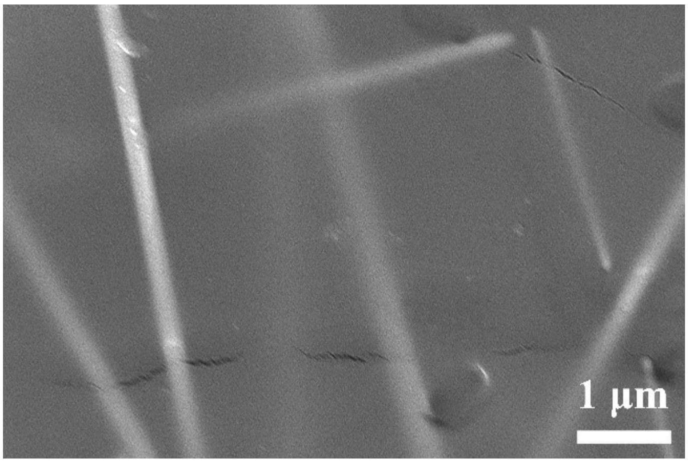


Figure S2 SEM image of the pristine Ag NW network embedded into PDMS supporting substrate

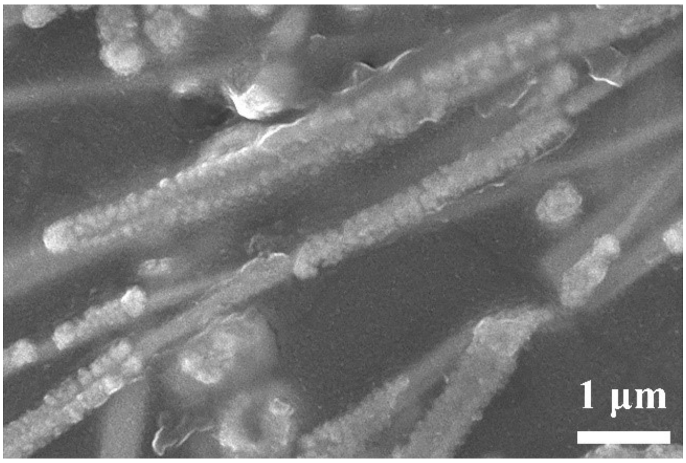


Figure S3 SEM image of Ag@NC NWs on PDMS substrate

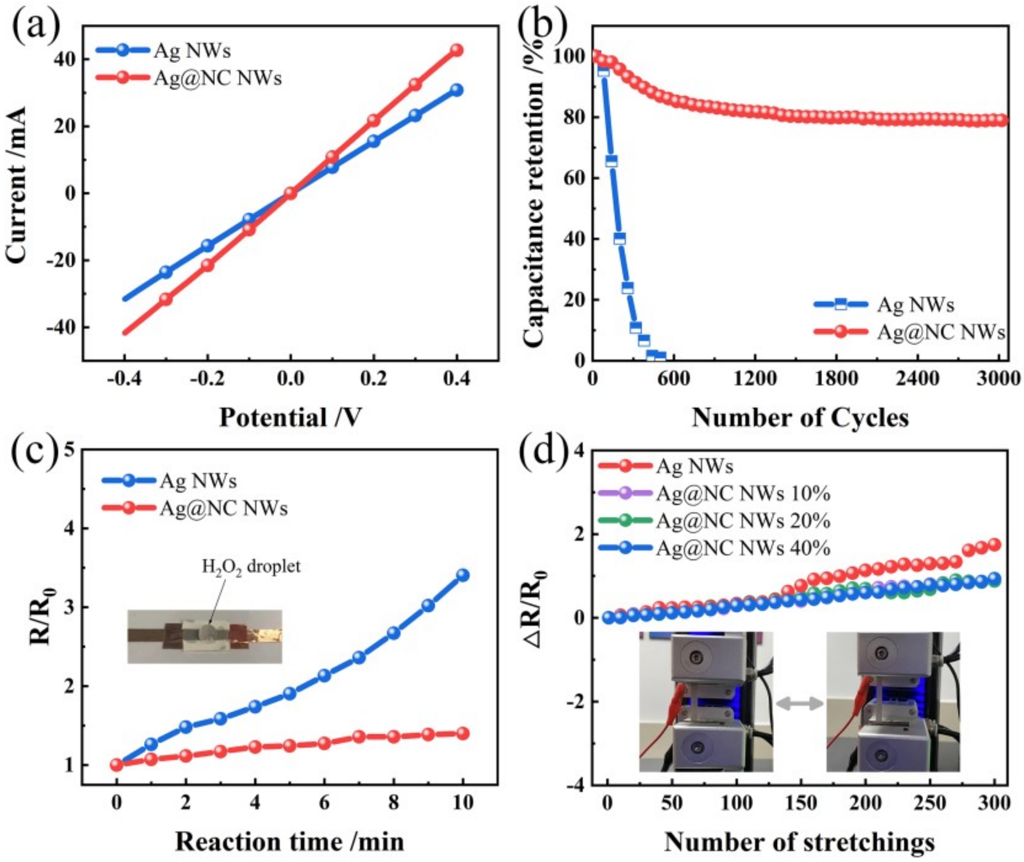


Figure S4 Investigation of Ag NW and Ag@NC NW networks on the conductivity, chemical stability, and mechanical stability. (a) I-V curves; (b) Capacity retention for 3,000 cycles at 200 mV $s^{-1}$; (c) Electrical resistance change under $H_2O_2$ exposure; (d) Resistance changes with different stretching lengths after 300 cycles.

The optimized Ag@NC NWs current collector has the following advantages: a) better electrical conductivity, improving 38% on the original basis. b) higher electrochemical stability with the capacitance retention of 80% after 3000 cycles, while the capacitance retention of the naked AgNWs loses entirely after several hundred cycles. c) higher corrosion resistance proved by a lower resistance change of the Ag@NC NW conductive networks under 30% $H_2O_2$ solution exposure. d) better tensile stability with resistivity changing less than 1.0 at 10, 20, and 40% stretching length.

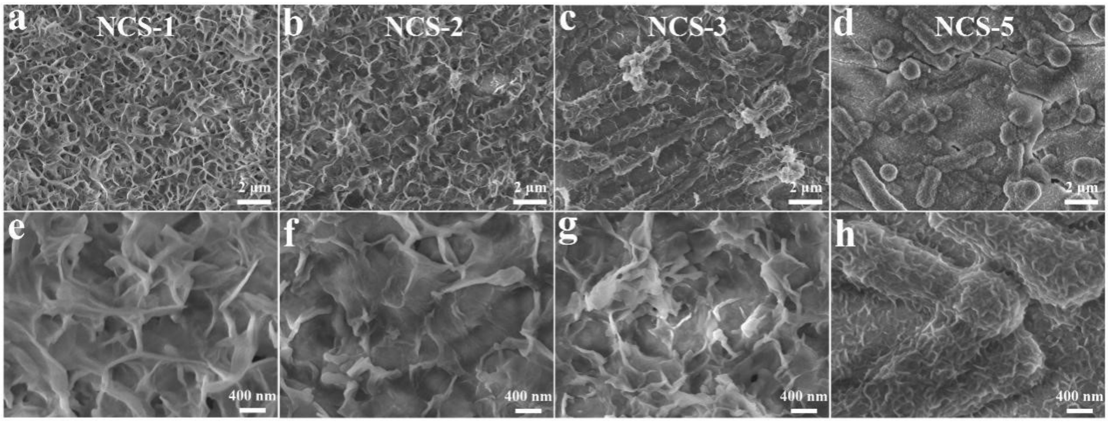


Figure S5 SEM images of (a) NCS-1; (b) NCS-2; (c)NCS-3; (d) NCS-5 electrodes with different magnifications

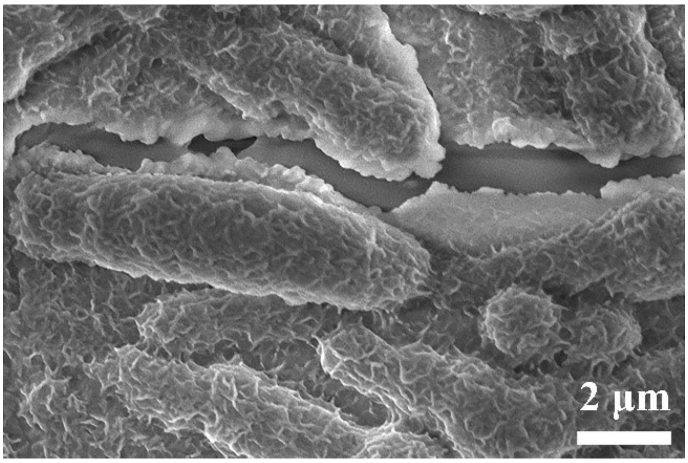


Figure S6 SEM image of NCS-5 electrode

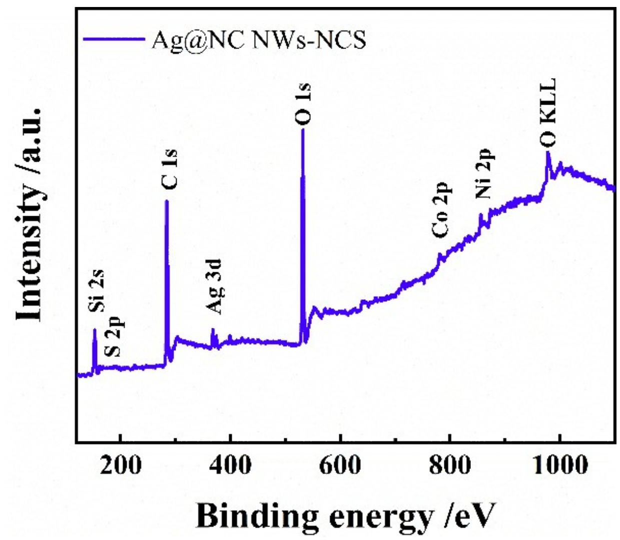


Figure S7 Full-range XPS spectra of the Ag@NC NWs-NCS electrode

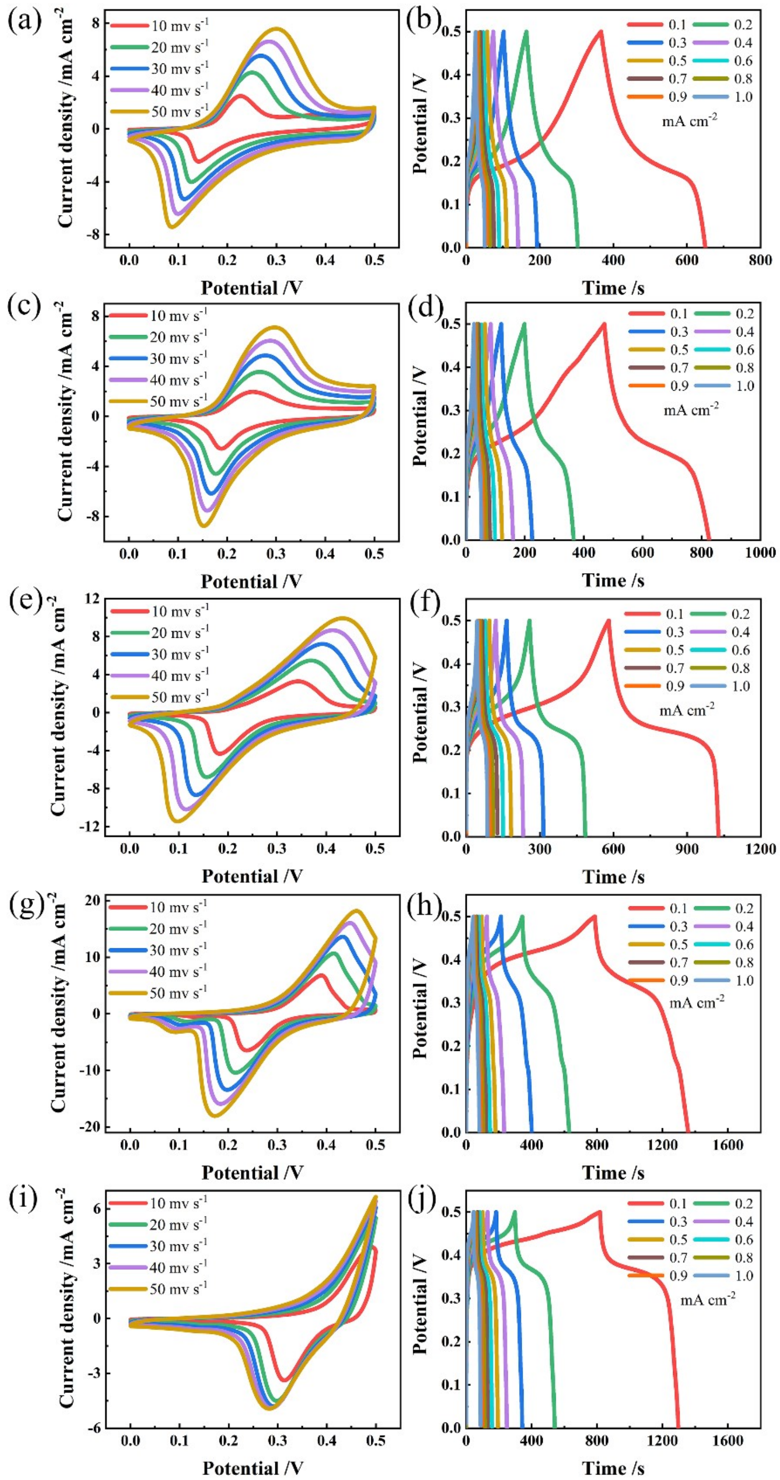


Figure S8 CV and GCD curves of the NCS-1, NCS-2, NCS-3, NCS-4, and NCS-5 electrodes

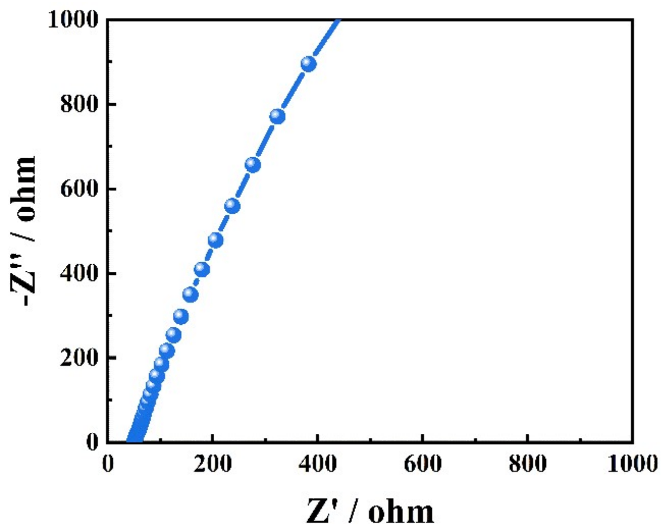


Figure S9 Nyquist plot of the supercapacitor based on the Ag@NC NWs-NCS and Ag@Bi/Al NWs electrodes

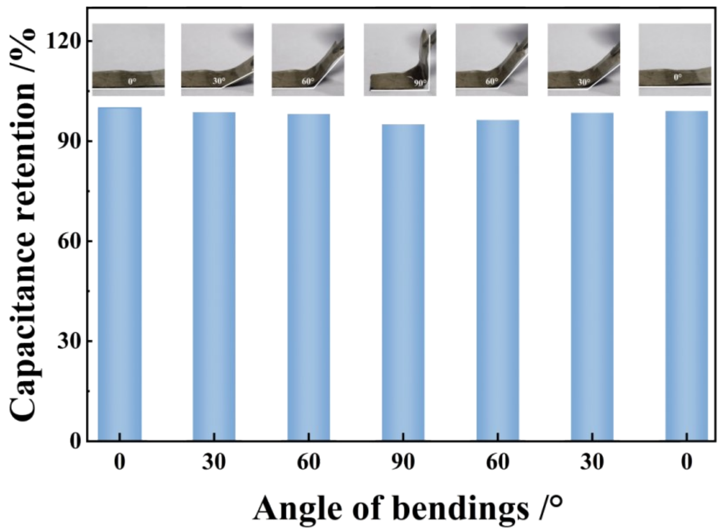


Figure S10 Capacitance retention rate of the device in various bent states

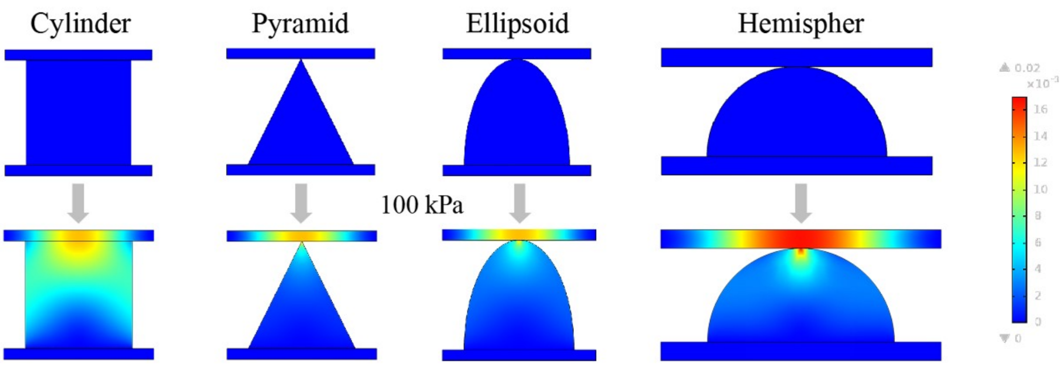


Figure S11 Deformability of different microstructured dielectric layers of PDMS under the pressure of 100 kPa.

We constructed different sensor models with the cylindrical, pyramidal, ellipsoidal, and hemispherical microstructures to explore a superior microstructure. When the sensors were loaded under 100 kPa, the hemispherical microstructure generated the most distinct change of *d* between the two electrodes.

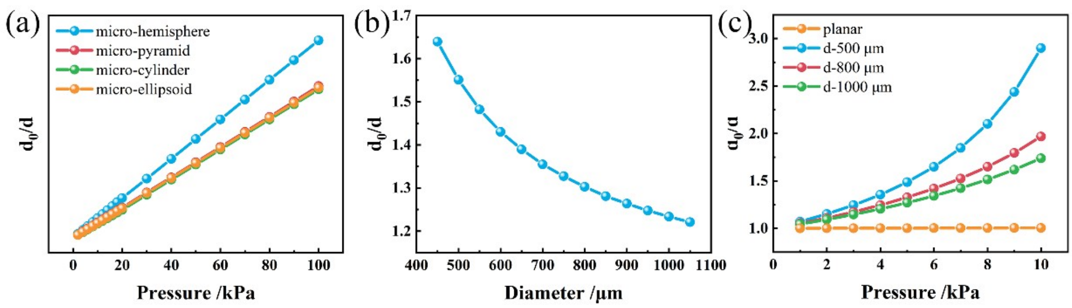


Figure S12 (a) Compression displacement comparison of sensors with different microstructured dielectric layers; Compression displacement of the micro-hemisphere dielectric layers under (b) different diameters and (c) different pressures.

The relationships between the electrode displacement and loading pressure for the microstructured sensors were further investigated by finite element analysis (FEA) in Fig. S12a, revealing the greatest deformability of the sensor with hemispherical microstructure. Moreover, we explored the effect of the hemisphere diameter on the displacement (Fig. S12b and S12c), indicating that the diameter of 500 μm allows the sensor to more significant deformation.

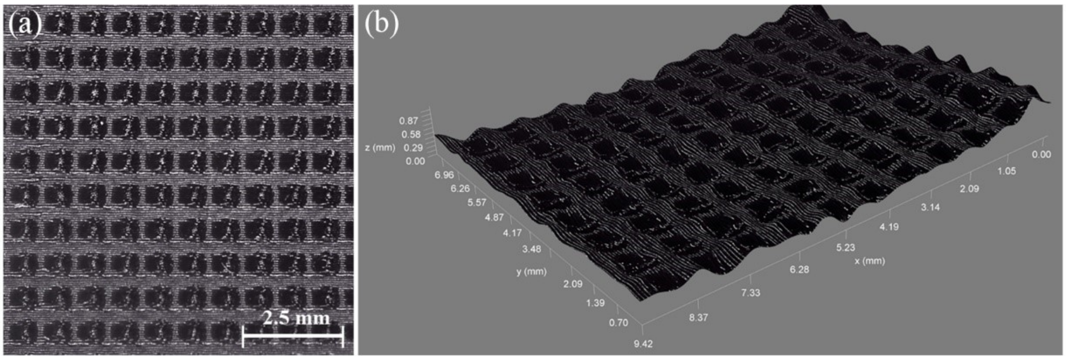


Figure S13 2D (a) and 3D Optical micrographs of the hemispherical antistructural template with a diameter of 500 μm

Table S1. Kinetic parameters of the NCS-1, NCS2, NCS-3, NCS-4, and NCS-5 electrodes fitted from Nyquist plots.

| Sample | Rs(Ω) | Rct(Ω) | CPE-T(Ω) | CPE-P(Ω) | W-R(Ω) | W-T(Ω) | W-P(Ω) |
|---|---|---|---|---|---|---|---|
| NCS-1 | 7.414 | 6.446 | $1.117\cdot10^{-7}$ | 0.985 | 13051 | 247.9 | 0.858 |
| NCS-2 | 9.701 | 16.630 | $8.877\cdot10^{-9}$ | 1.061 | 4.169 | 0.015 | 0.449 |
| NCS-3 | 7.059 | 8.143 | $1.681\cdot10^{-7}$ | 0.897 | 4.234 | 0.018 | 0.441 |
| NCS-4 | 6.584 | 3.642 | $4.511\cdot10^{-8}$ | 0.960 | 1.936 | 0.008 | 0.434 |
| NCS-5 | 7.412 | 4.422 | $2.502\cdot10^{-9}$ | 1.171 | 1.525 | 0.005 | 0.448 |

Table S2. Comparison of the other asymmetric supercapacitors and this work at energy density, power density, stability, etc.

| Materials | Energy density ($Wh\ kg^{-1}$) | Power Density ($W\ kg^{-1}$) | Stability (Cycles) | Flexible (Yes/No) | Current collector | Ref. |
|---|---|---|---|---|---|---|
| Ag@NC NWs-NCS //Ag@NC/BiAl NWs | 40.0 | 1100 | 91% (5000) | Yes 2000 (87.5%) | Ag@NC NWs | This work |
| $NiCo_2S_4/Co_8S_9$//AC | 33.5 | 150 | 78% (5000) | No | Ni foam | 1 |
| $NiCo_2S_4$/PRGO//AC | 27.5 | 446.5 | 80.1% (3000) | No | rGO black | 2 |
| $CuCo_2S_4$/GA//AC | 22 | 1080 | 84.5% (8000) | Yes | GO | 3 |
| $CoNi_2S_4$//AC | 33.9 | 409 | 78% (3000) | No | Ni foam | 4 |
| $NiMoS_4$@$NiS_2/MoS_2$//AC | 26.8 | 700 | 60% (5000) | No | carbon fiber paper | 5 |
| $NiCo_2S_4$@PPy-50/ NF//AC | 34.62 | 120.19 | 72.2% (3000) | No | Ni foam | 6 |
| $CuCo_2S_4$@$NiCo_2S_4$//AC | 23.4 | 400 | 100% (5000) | No | Ni foam | 7 |
| Co3$S_4$/$CoMo_2S_4$/rGO//AC | 33.1 | 850 | 97% (2000) | No | rGO | 8 |
| $Ni_3S_2$-CNTs//AC | 19.8 | 798 | 80% (1000) | No | CNT | 9 |
| $NiCo_2S_4$ NT@$NiCo_2S_4$ NSAs//rGO HSCs | 24.9 | 334 | 87.5% (5000) | No | Ni foam | 10 |

| | | | | | | |
|---|---|---|---|---|---|---|
| $NiCo_2S_4$ NTs//rGO HSCs | 31.5 | 156.6 | 92% (5000) | No | Ni foam | 11 |
| $NiCo_2O_4$/NiO//$Fe_2O_3$ | 19 | 157 | - | No | Ni foam | 12 |
| $KCu_7S_4$@NiMn LDH//AG | 15.9 | 800 | - | No | Ni foam | 13 |
| NiS-rGO//AC | 18.7 | 124 | - | No | Ni foam | 14 |
| NiCoS@SBA-C//SBA-C | 38.8 | 800 | 78.6% (6000) | No | Ni foam | 15 |
| AB-$NiCo_2S_4$//AC | 24.7 | 428 | - | No | Ni foam | 16 |
| $NiCo_2S_4$ nanoparticles//AC | 28.3 | 245 | 108% (1000) | No | Ni foam | 17 |
| $NiCo_2O_4$-rGO/AC | 23.32 | 324.9 | 91.6% (3000) | No | Ni foam | 18 |
| M $NiCo_2S_4$//AC | 25.5 | 334 | 93.4% (1500) | No | Ni foam | 19 |


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