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Supporting Information

Simple Route to Fiber-Shaped Heterojunctioned Nanocomposites for Knittable High-Performance Supercapacitors

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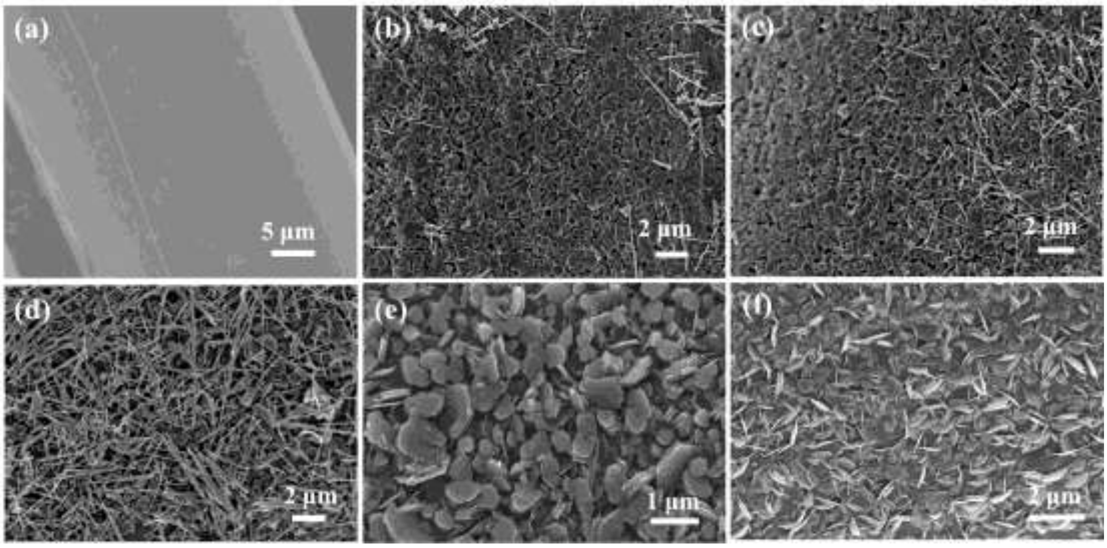

Fig. S1 SEM images of (a) pure stainless steel microwire (D = 30 μm), and (b-d) the $NiCo_2S_4$@MgS nanocomposites synthesized via the first-step hydrothermal reaction in the solution containing $NiCl_2 \cdot 6H_2O$, $CoCl_2 \cdot 6H_2O$, $MgCl_2 \cdot 6H_2O$, urea and NH_4F at 120 °C for different times: (b) 12 h, (c) 16 h and (d) 20 h, followed by the second-step hydrothermal reaction in the Na_2S solution at 80 °C for 6 h. (e) SEM image of the $NiCo_2S_4$ nanosheets grown on the stainless steel microwire prepared via the first-step hydrothermal reaction in the solution containing only $NiCl_2 \cdot 6H_2O$, $CoCl_2 \cdot 6H_2O$, urea and NH_4F (i.e., no $MgCl_2 \cdot 6H_2O$) at 120 °C for 24 h, followed by the second-step hydrothermal reaction in the Na_2S solution at 80 °C for 6 h. (f) SEM image of the MgS film grown on the stainless steel wire produced via the first-step hydrothermal reaction in the solution containing solely $MgCl_2 \cdot 6H_2O$, urea and NH_4F (i.e., no $NiCl_2 \cdot 6H_2O$, $CoCl_2 \cdot 6H_2O$) at 120 °C for 24 h, followed by the second-step hydrothermal reaction in the Na_2S solution at 80 °C for 6 h.

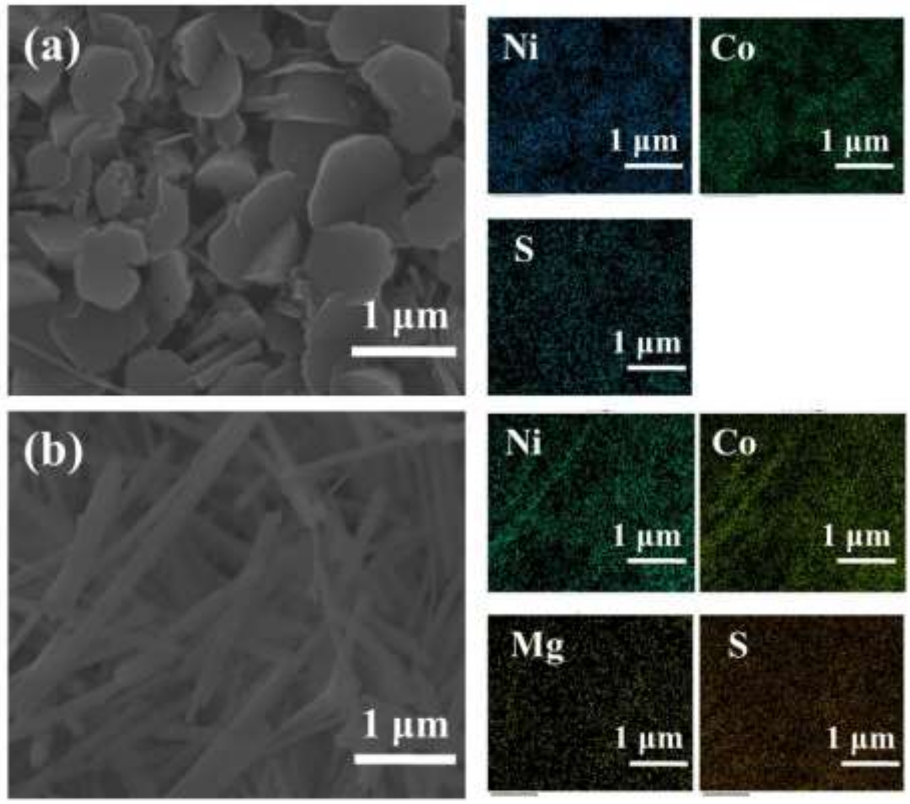

Fig. S2 EDS mapping of the (a) $NiCo_2S_4$ nanosheets and (b) MgS nanowires within the $NiCo_2S_4$@MgS nanocomposite.

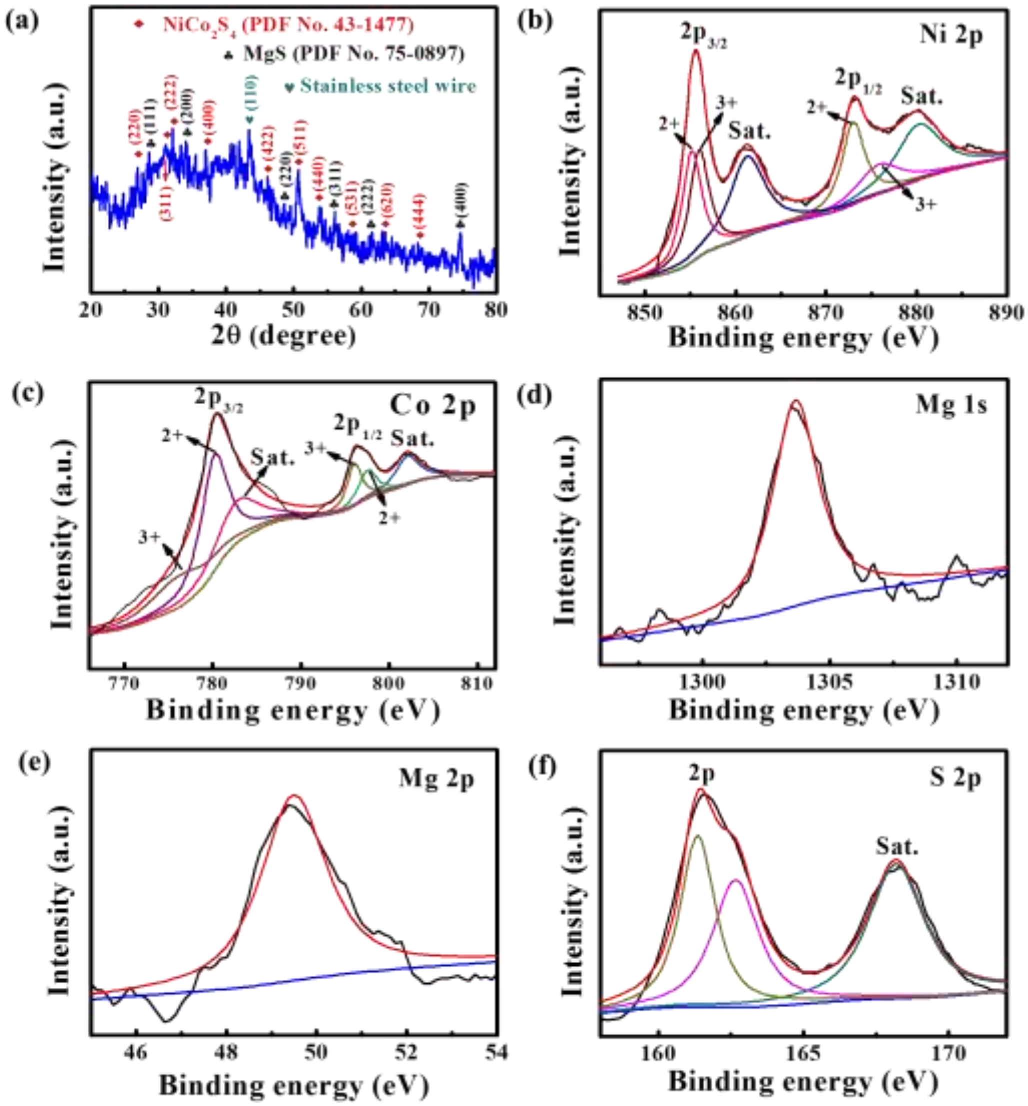

Fig. S3 (a) XRD pattern of the $NiCo_2S_4$@MgS nanocomposites. High-resolution XPS spectra for the (b) Ni 2p, (c) Co 2p, (d) Mg 1s, (e) Mg 2p and (f) S 2p in the $NiCo_2S_4$@MgS nanocomposites.

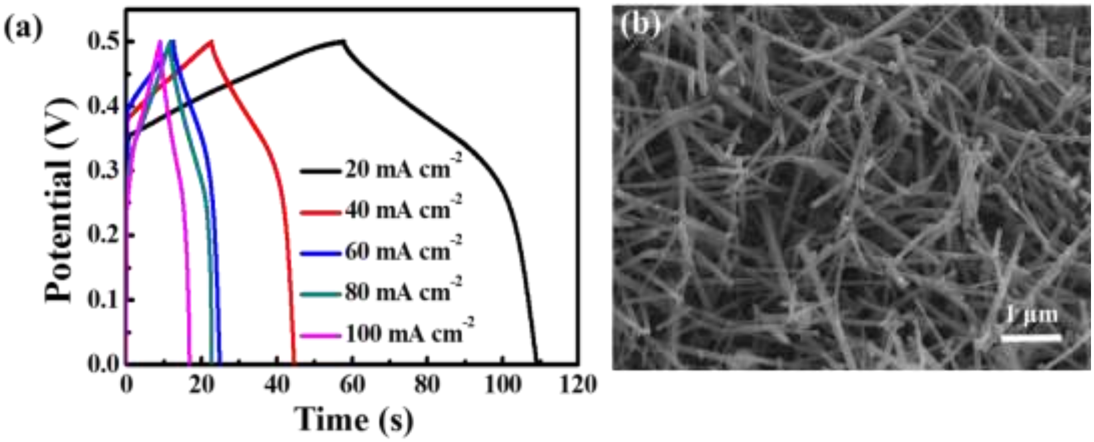

Fig. S4 (a) Galvanostatic charge-discharge (GCD) of the $NiCo_2S_4$@MgS electrode at high current densities. (b) SEM image of the $NiCo_2S_4$@MgS electrode after the charge-discharge 10000-cycling test.

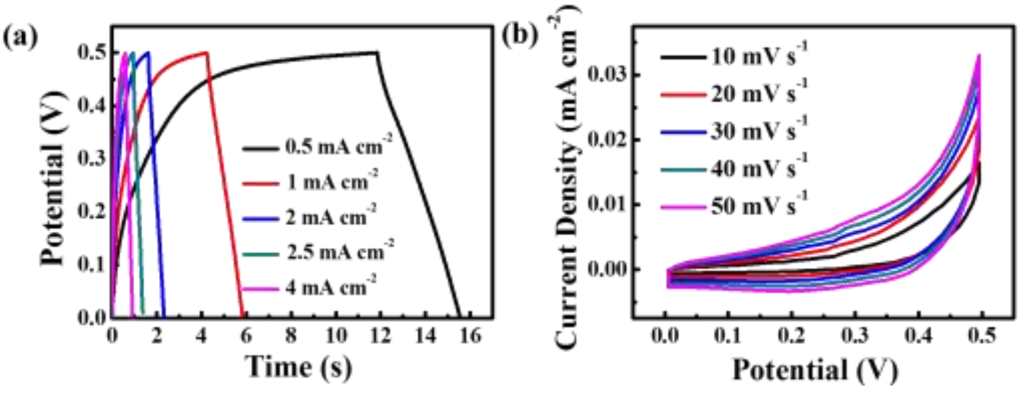

Fig. S5 (a) GCD and (b) CV curves of the MgS film on the stainless steel microwire prepared via the first-step hydrothermal reaction in the solution only containing $MgCl_2 \cdot 6H_2O$, urea and NH_4F at 120 °C for 24 h, followed by the second-step hydrothermal reaction in the Na_2S solution at 80 °C for 6 h.

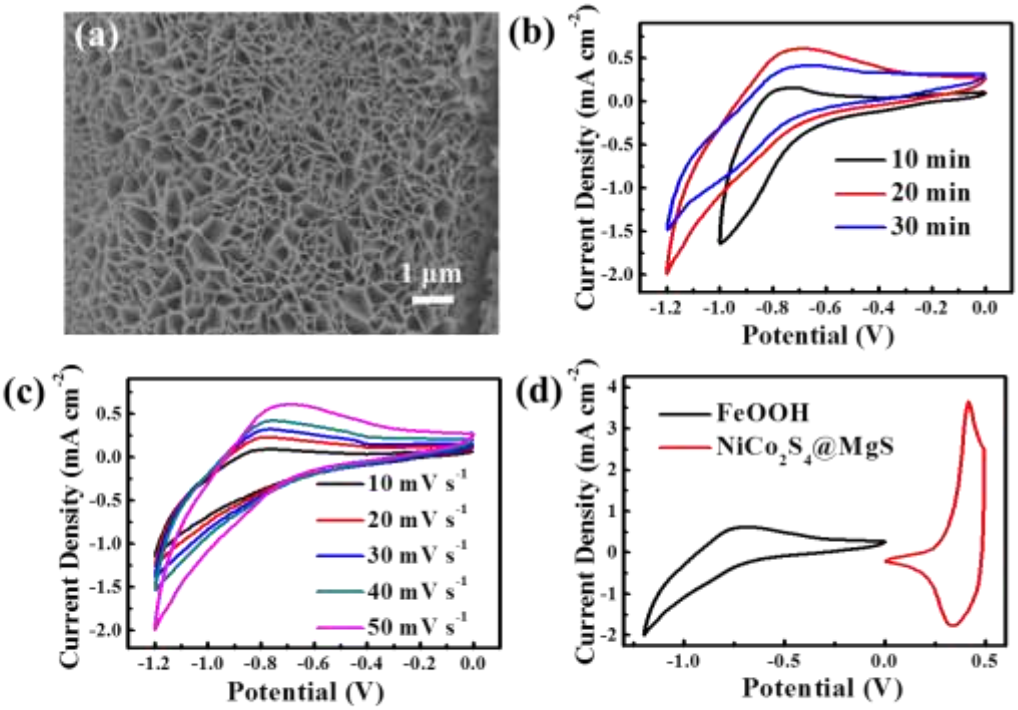

Fig. S6 (a) SEM image of the FeOOH electrode prepared at a deposition time of 20 min. (b) CV curves at a scan rate of 50 mV s^{-1} of the FeOOH electrodes grown on stainless steel microhairs via the electrodeposition method at different deposition times. (c) CV curves at different scan rates of the FeOOH electrode prepared for a deposition time of 20 min. (d) CV curves at a scan rate of 50 mV s^{-1} of the $NiCo_2S_4$@MgS and FeOOH electrodes prepared for a deposition time of 20 min.

In order to assemble fiber-shaped asymmetric supercapacitors containing the $NiCo_2S_4$@MgS nanocomposites as the positive electrode, the FeOOH negative electrode (**Figure S5a**) was grown on stainless steel microwire by electrodeposition. **Figure S5b** shows the CV curves of the FeOOH negative electrode prepared at different deposition times. It can be seen that the sample obtained at the deposition time of 20 min has the best CV performance and its CV curves tested at different scan rate are shown in **Figure S5c**. The reactions of the FeOOH electrode are as follows:[1]

$$NO^{3-} + 7H_2O + 8e^- = NH_4^+ + 10OH^-$$

$$Fe^{3+} + 3OH^- = Fe(OH)_3$$

$$Fe(OH)_3 = FeOOH + H_2O$$

The CV curves of the FeOOH and $NiCo_2S_4$@MgS electrodes are shown in **Figure S5d**. The area of the CV curve suggests the quantity of electric charge (Q) of the electrodes, and the area ratio of $NiCo_2S_4$@MgS to FeOOH is 1:0.8, indicating that the Q ratio is 1:0.8.

Table S1. Comparison of the volumetric energy density (*E*) and power density (*P*) between the FASCs in the present study and the reported values in literatures.

Samples (positive electrode//negative electrode)	***E* (mWh cm^{-3})**	***P* (W cm^{-3})**	***Refs.***
$NiCo_2S_4$@MgS network-like nanostructures on stainless steel microhairs//FeOOH nanosheets on stainless steel microhairs	107.5	1.7	This work
$NiCo_2O_4$ nanosheets on Ni wires//$NiCo_2O_4$ nanosheets on Ni wires	1.44	17	25
carbon fiber thread@PANI//functionalized carbon fiber thread	2	10.22	45
graphene nanoribbon fiber//MnO_2 on graphene nanoribbon fiber	5.7	0.08	46
$CoNiO_2$ nanowires on carbon fiber//active carbon on carbon fiber	0.50	0.213	47
porous Mn-Ni-Co-S nanotubes on carbon nanotube fiber//VN nanowires on carbon nanotube fiber	52	0.582	48
Fe_2O_3 on carbon fiber//MnO_2 on carbon nanotube-web paper	0.43	0.02	49
MnO_2 on carbon fiber//FeOOH/PPy on carbon fiber	3.7	0.19	50
Mo-Ni-Co-O nanowire on carbon nanotube	22.2	0.21	51

fiber//VN nanowires on carbon nanotube fiber			
Zn-Ni-Co-O nanowires on carbon nanotube fiber//VN@C on carbon nanotube strip	33.66	0.40	52
PANI@CNTs on carbon fiber//MnO_2@CNTs on carbon fiber	8.3	0.1	53