Supplementary Information

3D-Printed Stretchable Structural Supercapacitor with Active Stretchability/Flexibility and Remarkable Volumetric Capacitance

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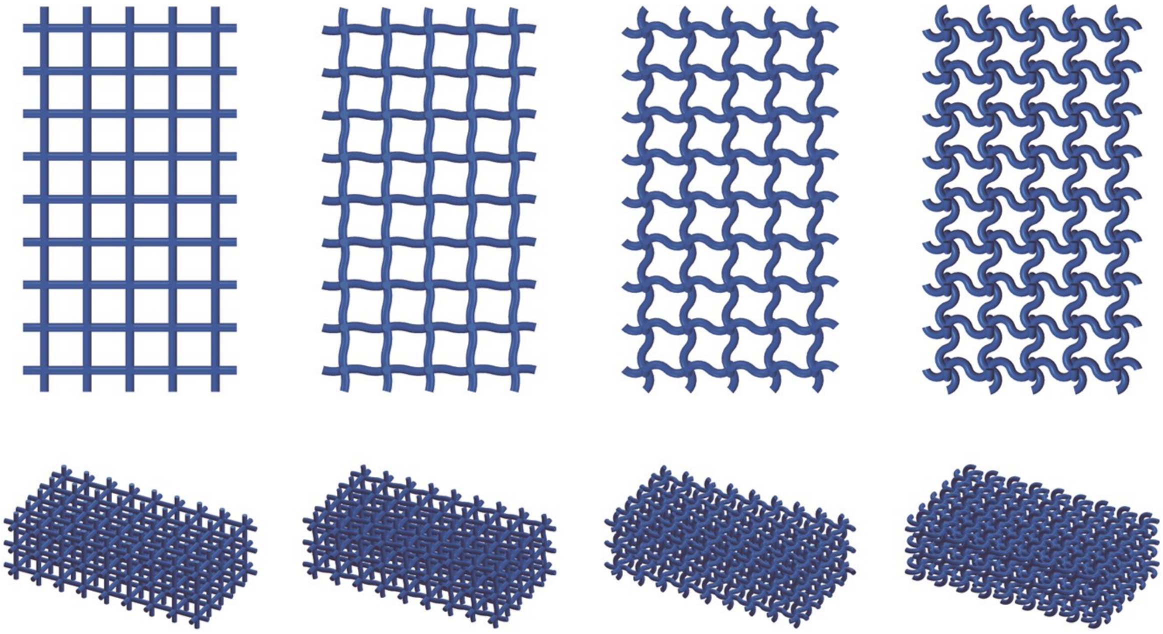

Fig. S1. Negative Poisson's ratio structures with different concave angles (From left to right: 0°, 30°, 90°, and 150°).

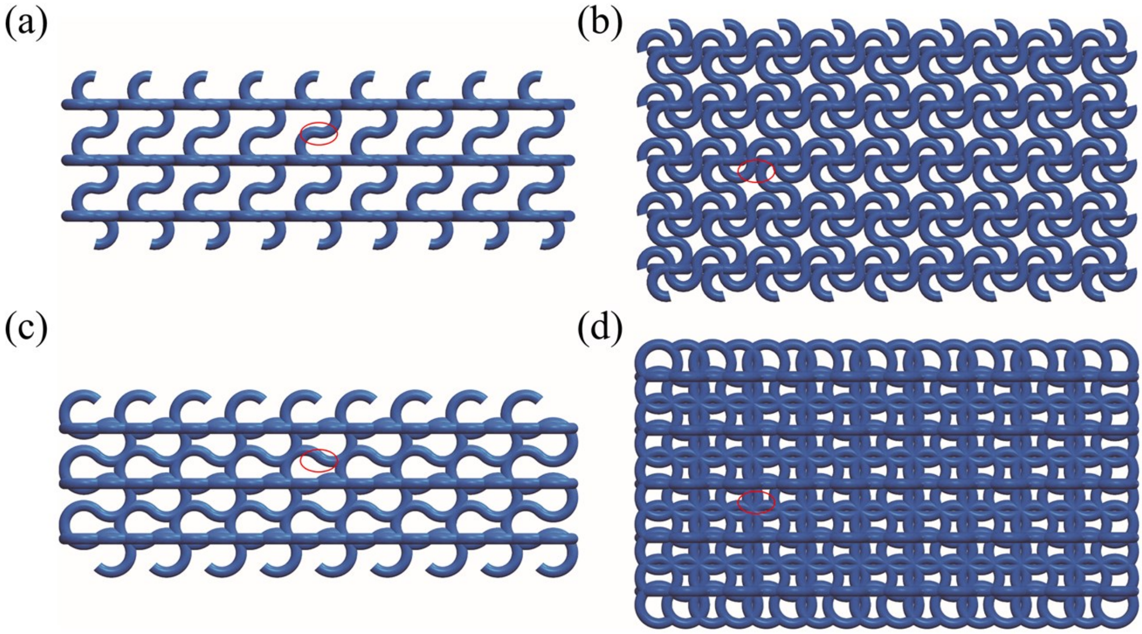

Fig. S2. Side view (a, c) and top view (b, d) of Negative Poisson's ratio structures with concave angles of 210° (up) and 270° (down).

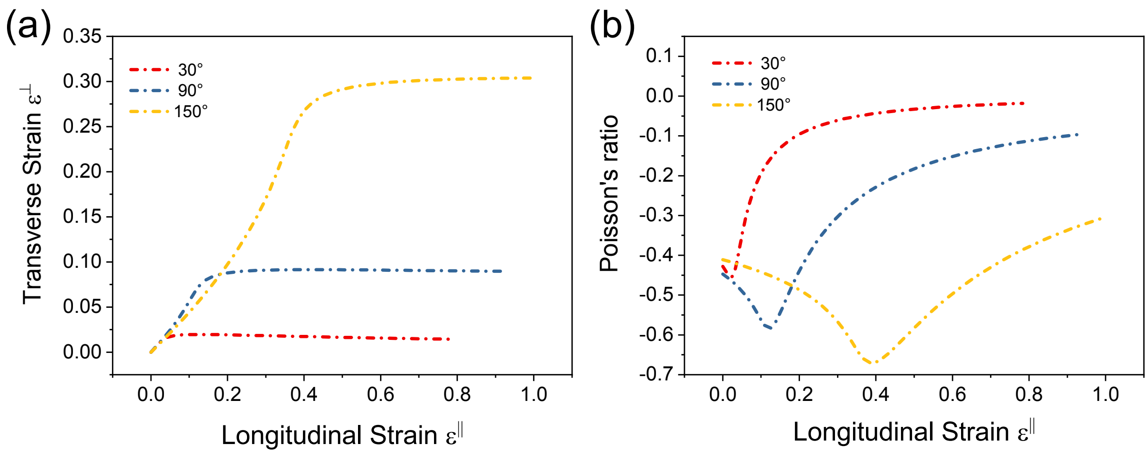

Fig. S3. Longitudinal strain vs transverse strain (a) and Poisson's ratio (b) of Negative Poisson's ratio structures with concave angles of 30°, 90° and 150°.

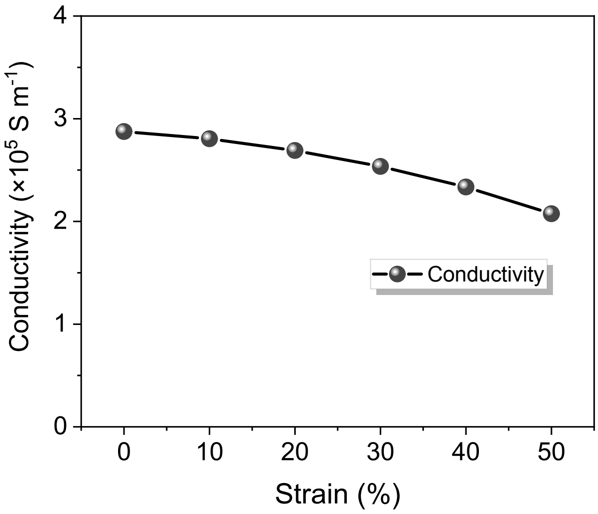

Fig. S4. Conductivity of $CoNi_2S_4$/NiCo-LDHs/Ni/PL-150° under different tensile strains.

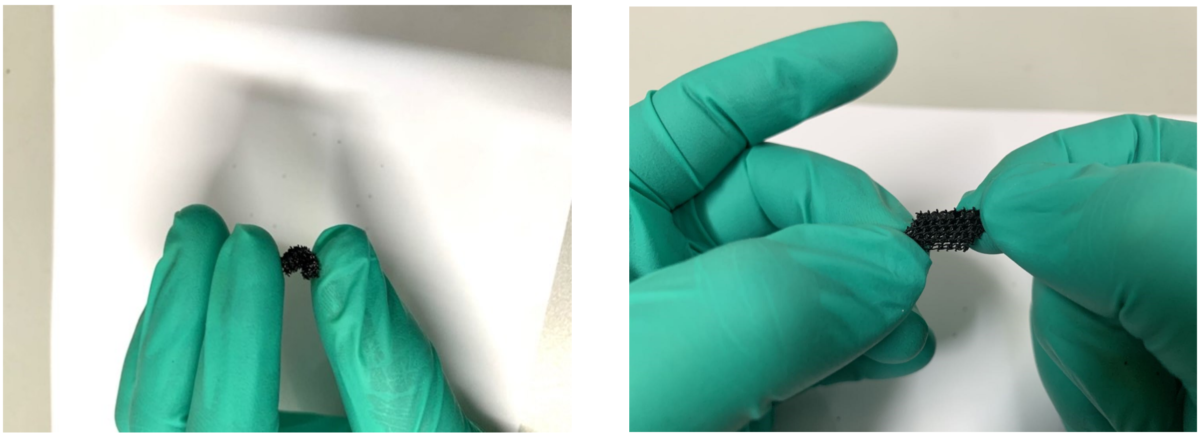

Fig. S5. The photographs of electrodes during the stretching/bending process

To analyze the deformation of the designed structures during the stretching process, we conducted finite element simulation by using the Comsol Multiphasics. We used the viscoelastic model to capture the mechanical behavior of cured photosensitive resin. The generalized Maxwell model was used and the applied strain rate was 0.02 mm s^{-1}. Without considering the thermal effect, the density, modulus and Poisson's ratio of the cured photosensitive resin were 1.3 g cm^{-3}, 2580 MPa and 0.35, respectively (at room temperature). The simulation result was shown in Movie. S1.

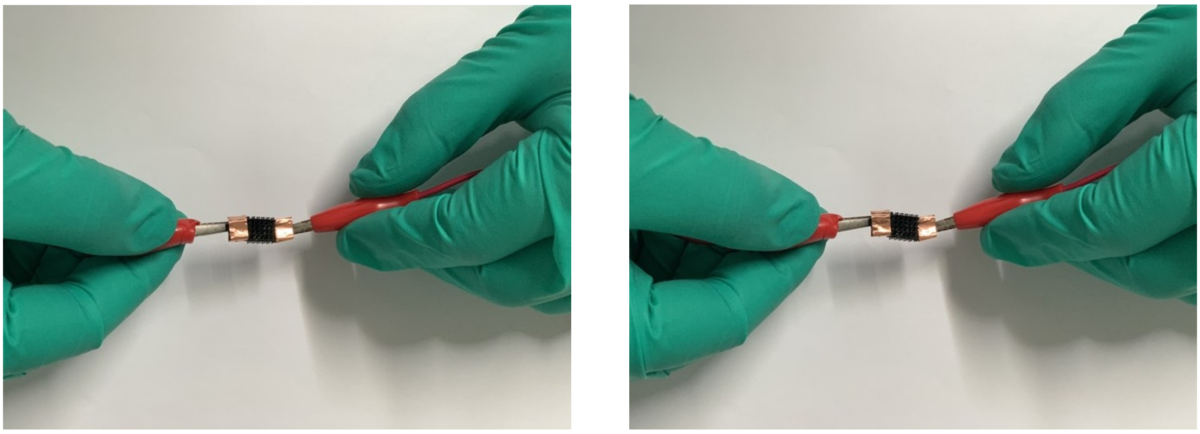

Fig. S6. The photographs of the assembled symmetric supercapacitor.

Table S1 Atomic and mass concentration of the elements in $CoNi_2S_4$/NiCo-LDHs.

Atomic Concentration %					Mass Concentration %				
Ni 2p	Co 2p	O 1s	C 1s	S 2p	Ni 2p	Co 2p	O 1s	C 1s	S 2p
7.94	9.68	37.71	39.54	5.12	20.45	25.04	26.47	20.83	7.2

Table S2 Comparison of the 3D-printed stretchable structural $CoNi_2S_4$/NiCo-LDHs/Ni/PL nanocomposite electrodes with recently reported 3D electrodes in terms of volumetric capacitance, rate capability and stability.

Electrode Materials	Volumetric Capacitance	Rate Capability	Stability	Ref.
PPy-PVA hydrogel	13.06 F cm^{-3} at 5 mA cm^{-3}	15.8% at 50 mA cm^{-3}	86.3% after 10000 cycles	43
AC/CNT/rGO	41.3 F cm^{-3} at 10 mV s^{-1}	72% at 100 mV s^{-1}	93.3% after 5000 cycles	41
$CoNi_2S_4$/Ni	3.17 F cm^{-3} at 10 mA cm^{-3}	68.5% at 50 mA cm^{-3}	77.3% after 2000 cycles	24
PANI/rGO	12.5 F cm^{-3} at 4.2 mA cm^{-2}	~50% at 50 mA cm^{-2}	75% after 10000 cycles	37
Black-Phosphorus/CNT	41.1 F cm^{-3} at 5 mV s^{-1}	83.7% at 10 mV s^{-1}	91.5% after 10000 cycles	42
ZNCO@$Ni(OH)_2$ NWAs/CNTF	94.67 F cm^{-3} at 3 mA cm^{-2}	74% at 15 mA cm^{-2}	90.3% after 10000 cycles	38
MnO_2/MGF/NF	14.28 F cm^{-3} at 0.64 mA cm^{-2}	66.1% at 10.24 mA cm^{-2}	~100% after 5000 cycles	39
$Cu(OH)_2$/Cu/Ceramic	8.46 F cm^{-3} at 5 mA cm^{-3}	68.1% at 1 A cm^{-3}	84.3% after 10000 cycles	17
rGO/CNT	2 F cm^{-3} at 0.1 mA cm^{-2}	84% at 1 mA cm^{-2}	90% after 100000 cycles	40
$CoNi_2S_4$/NiCo-LDHs/Ni/PL	46.2 F cm^{-3} at 10 mA cm^{-3}	76.8% at 500 mA cm^{-3}	92.2 after 5000 cycles	**This work**

Table S3 Comparison of the 3D-printed stretchable structural $CoNi_2S_4$/NiCo-LDHs/Ni/PL supercapacitor with state-of-the-art flexible supercapacitors in terms of capacitance retention after stretching/bending cycles.

Electrode Materials	Capacitance Retention After Bending Cycles	Capacitance Retention After Stretching Cycles	Ref.
PPy−PVA hydrogel	97.9% after 10000 cycles	\	43
PPy/BPO-CNT	\	95% after 10000 cycles	9
CNT-MnO_2	80.8 % after 1800 cycles	\	13
PANI/RGO	\	~105% of after 500 cycles	10
Ni_3S_2-Ni wire	96% after 50 cycles	\	54
MXene-AgNW-MnONW-C60 gel	\	75% after 1000 cycles	8
ZNCO@$Ni(OH)_2$ NWAs/CNTF	93.6% after 3000 cycles	\	38
Graphene/Nanotube /PANI	\	93% after 100 cycles	7
SE/rGO/Ag-NWs/rGO/ SE	97% after 4000 cycles	99% after 1000 cycles	14
$CoNi_2S_4$/NiCo-LDHs/Ni/PL	75.2% after 1000 cycles	88.1% after 2000 cycles	**This work**

Table S4 Statistical results in tensile strength, modulus, energy absorption, and designed surface area of four types of electrodes.

Sample	Tensile Strength (MPa)	Modulus (MPa)	Energy Absorption (kJ m^{-3})	Designed Surface Area (cm^2)
0°	1.77 ± 0.18	15.92 ± 1.21	133.62 ± 17.88	6.82
30°	1.86 ± 0.11	10.95 ± 1.64	349.52 ± 29.40	8.74
90°	2.07 ± 0.15	8.56 ± 1.57	505.21 ± 39.55	9.50
150°	2.21 ± 0.23	6.88 ± 0.75	817.73 ± 58.91	11.39