

# Doping-driven electronic structure and conductivity modification of nickel sulfide

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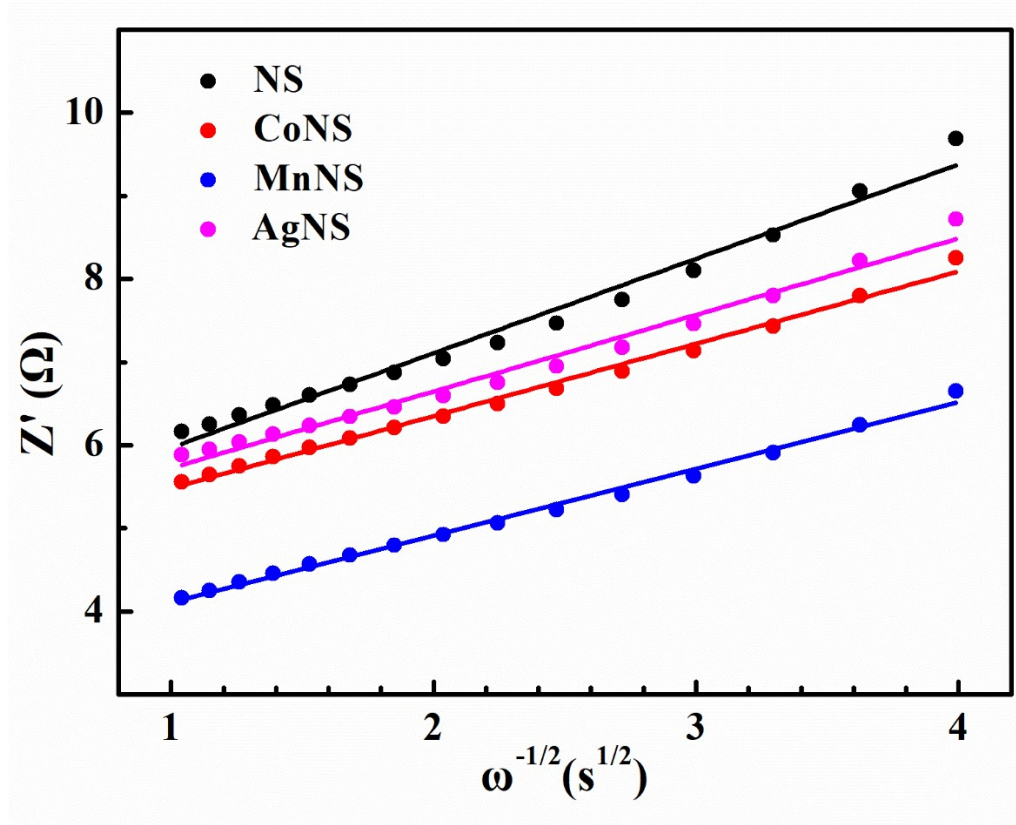
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1. Relationship between real impedance and  $\omega^{-1/2}$  of NS, CoNS, MnNS and AgNS Electrode in the low-frequency region is liner fitted.



**Fig. S1** Relationship between real impedance and reciprocal square root of lower angular frequencies

2. Dopant concentration in synthesized samples are detected by ICP-AES.

**Table S1** Doped samples composition from ICP-AES

Sample	2% CoNS		0.5% MnNS		0.5% AgNS	
Element	Ni	Co	Ni	Mn	Ni	Ag
Mass fraction(%)	40.88%	1.02%	26.14%	0.09%	23.12%	0.23%

Atomic ratio of transition metals: Ni(%)	2.48%	0.36%	0.54%
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3. Ni 2P binding energy of fitted peaks is shown below.

**Table S2** Ni 2P binding energy of fitted peaks

Sample	Ni 2P <sub>3/2</sub>			Ni 2P <sub>1/2</sub>		
	Ni <sup>2+</sup>	Ni <sup>3+</sup>	Sat.	Ni <sup>2+</sup>	Ni <sup>3+</sup>	Sat.
NS	853.43 eV	856.60 eV	860.85 eV	870.49 eV	874.30 eV	879.33 eV
2% CoNS	853.86 eV	856.51 eV	860.61 eV	871.46 eV	874.66 eV	879.24 eV
0.5% MnNS	854.19 eV	856.45 eV	860.51 eV	871.62 eV	874.87 eV	879.58 eV
0.5% AgNS	853.98 eV	856.12 eV	860.53 eV	871.54 eV	874.70 eV	879.32 eV

4. The parameters of model in the equivalent circuit are simulated and listed in Table S3.

**Table S3** Kinetic Parameters of NS, CoNS, MnNS and AgNS Electrode

Sample	R <sub>s</sub>	R	R <sub>ct</sub>	σ <sub>w</sub>	D <sub>K</sub> <sup>+</sup>
NS	0.12 Ω	0.18 Ω	4.93 Ω	1.14 Ω s <sup>-0.5</sup>	2.77×10 <sup>-12</sup> cm <sup>2</sup> ·s <sup>-1</sup>
2% CoNS	0.11 Ω	0.16 Ω	4.45 Ω	0.87 Ω s <sup>-0.5</sup>	4.72×10 <sup>-12</sup> cm <sup>2</sup> ·s <sup>-1</sup>
0.5% MnNS	0.10 Ω	0.18 Ω	3.19 Ω	0.80 Ω s <sup>-0.5</sup>	5.56×10 <sup>-12</sup> cm <sup>2</sup> ·s <sup>-1</sup>
0.5% AgNS	0.12 Ω	0.22 Ω	4.79 Ω	0.92 Ω s <sup>-0.5</sup>	4.21×10 <sup>-12</sup> cm <sup>2</sup> ·s <sup>-1</sup>

5. Comparison of specific capacitance in this work and other previous reports.

**Table S4** Comparison of specific capacitance with similar nickel sulfides

Samples	Specific capacitance	Current density	Electrolyte
PN-rGO/NCS <sup>1</sup>	1687 F g <sup>-1</sup>	0.5 A g <sup>-1</sup>	6 M KOH
SnNi <sub>2</sub> S <sub>4</sub> <sup>2</sup>	1483.8 F g <sup>-1</sup>	2 A g <sup>-1</sup>	2 M KOH
Hollow sphere NiS <sub>2</sub> <sup>3</sup>	1382.0 F g <sup>-1</sup>	1 A g <sup>-1</sup>	2 M KOH
NiS <sub>2</sub> <sup>4</sup>	695 F g <sup>-1</sup>	1.25 A g <sup>-1</sup>	3 M KOH
NiS <sub>2</sub> /ZnS <sup>5</sup>	1198 F g <sup>-1</sup>	1 A g <sup>-1</sup>	3 M KOH
NiS-NiCo <sub>2</sub> O <sub>4</sub> @C <sup>6</sup>	1411 F g <sup>-1</sup>	1 A g <sup>-1</sup>	6 M KOH
C@MoS <sub>2</sub> /Ni <sub>3</sub> S <sub>4</sub> <sup>7</sup>	951.3 F g <sup>-1</sup>	2 A g <sup>-1</sup>	2 M KOH
NiS/ACNTs <sup>8</sup>	1266 F g <sup>-1</sup>	1 A g <sup>-1</sup>	3 M KOH
Co-NiS/NCDs <sup>9</sup>	2480 F g <sup>-1</sup>	1 A g <sup>-1</sup>	3 M KOH
graphene/NiS <sub>2</sub>	478.1 F g <sup>-1</sup>	0.5 A g <sup>-1</sup>	6 M KOH
NS	2221.4 F g <sup>-1</sup>	1 A g <sup>-1</sup>	2 M KOH
2% CoNS	2874.6 F g <sup>-1</sup>	1 A g <sup>-1</sup>	2 M KOH
0.5% MnNS	2612.5 F g <sup>-1</sup>	1 A g <sup>-1</sup>	2 M KOH
0.5% AgNS	2587.3 F g <sup>-1</sup>	1 A g <sup>-1</sup>	2 M KOH

6. Comparison of energy density as asymmetric supercapacitor in this work and other previous reports.

**Table S5** Comparison of energy density asymmetric supercapacitor with similar nickel sulfides

Sample	Energy density	Power density
N2//N2 <sup>10</sup>	9.00 Wh Kg <sup>-1</sup>	233.0 W Kg <sup>-1</sup>
NiCo2S4/CC//AC/CC <sup>11</sup>	25.2 Wh Kg <sup>-1</sup>	799.6 W Kg <sup>-1</sup>
Ni-Co LDH/STSC-0-800//a-STSC-1-600 <sup>12</sup>	23.5 Wh Kg <sup>-1</sup>	959.7 W Kg <sup>-1</sup>
Ni/Co MOF//AC <sup>13</sup>	12.8 Wh Kg <sup>-1</sup>	372.5 W Kg <sup>-1</sup>
NCO//AC <sup>14</sup>	15.8 Wh Kg <sup>-1</sup>	1385 W Kg <sup>-1</sup>
MoS2/NiS//AC <sup>15</sup>	9.80 Wh Kg <sup>-1</sup>	1524 W Kg <sup>-1</sup>
NS//rGO(this work)	20.8 Wh Kg <sup>-1</sup>	800 W Kg <sup>-1</sup>
	17.5 Wh Kg <sup>-1</sup>	1600 W Kg <sup>-1</sup>
	10.0 Wh Kg <sup>-1</sup>	4000 W Kg <sup>-1</sup>
	6.40 Wh Kg <sup>-1</sup>	6400 W Kg <sup>-1</sup>
	5.33 Wh Kg <sup>-1</sup>	8000 W Kg <sup>-1</sup>
2% CoNS//rGO(this work)	36.6 Wh Kg <sup>-1</sup>	800 W Kg <sup>-1</sup>
	31.6 Wh Kg <sup>-1</sup>	1600 W Kg <sup>-1</sup>
	20.6 Wh Kg <sup>-1</sup>	4000 W Kg <sup>-1</sup>
	13.2 Wh Kg <sup>-1</sup>	6400 W Kg <sup>-1</sup>
	10.0 Wh Kg <sup>-1</sup>	8000 W Kg <sup>-1</sup>
0.5% MnNS//rGO(this work)	36.1 Wh Kg <sup>-1</sup>	800 W Kg <sup>-1</sup>

	31.5 Wh Kg <sup>-1</sup>	1600 W Kg <sup>-1</sup>
	18.9 Wh Kg <sup>-1</sup>	4000 W Kg <sup>-1</sup>
	12.4 Wh Kg <sup>-1</sup>	6400 W Kg <sup>-1</sup>
	9.11 Wh Kg <sup>-1</sup>	8000 W Kg <sup>-1</sup>
0.5% AgNS//rGO(this work)	36.0 Wh Kg <sup>-1</sup>	800 W Kg <sup>-1</sup>
	30.3 Wh Kg <sup>-1</sup>	1600 W Kg <sup>-1</sup>
	17.7 Wh Kg <sup>-1</sup>	4000 W Kg <sup>-1</sup>
	11.2 Wh Kg <sup>-1</sup>	6400 W Kg <sup>-1</sup>
	8.67 Wh Kg <sup>-1</sup>	8000 W Kg <sup>-1</sup>

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