

**Supporting Information**

**Novel Ordered Hollow Spherical Nickel Silicate-Nickel Hydroxide United Composite with Two  
Types of Morphologies for Enhanced Electrochemical Storage Performance**

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### ***Preparation of Ni(OH)<sub>2</sub>***

In a typical synthesis of the layered Ni(OH)<sub>2</sub>, 5.8 mmol Ni(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O was dissolved in 18 mL deionized water under constant stirring. Then, 5 mmol NH<sub>4</sub>Cl and 1 mL NH<sub>3</sub>·H<sub>2</sub>O was dissolved in 20 mL deionized water under constant stirring. After the solutions are mixed, a further stirring for 30 min is in process. The final mixture was transferred into a 50 mL Teflon-lined stainless autoclave and treated at 120 °C for 20 h. The result product was cooled down to room temperature and collected by filtration, then washed with water and ethanol three times repeatedly. Finally, the product was dried at 60 °C for 12 h before used.

### ***Preparation of NiSiO<sub>x</sub>***

0.1 g SiO<sub>2</sub> sphere was dispersed into 18 mL deionized by ultrasonication. 3.7 mmol NiCl<sub>2</sub>·6H<sub>2</sub>O, 12 mmol NH<sub>4</sub>Cl, 0.24 mL NH<sub>3</sub>·H<sub>2</sub>O was added into 20 mL deionized water under vigorously stirring. The two solutions were mixed and continuous stirred for another 10 min. Then, a hydrothermal treatment was carried by transferred the mixture into a Teflon-lined stainless-steel autoclave and heated at 120 °C for 20 h. Finally, the products were collected by centrifugation and washed with deionized water and ethanol 3 times repeatedly, along with vacuum dried at 60 °C for 12 h.

**Figure S1**

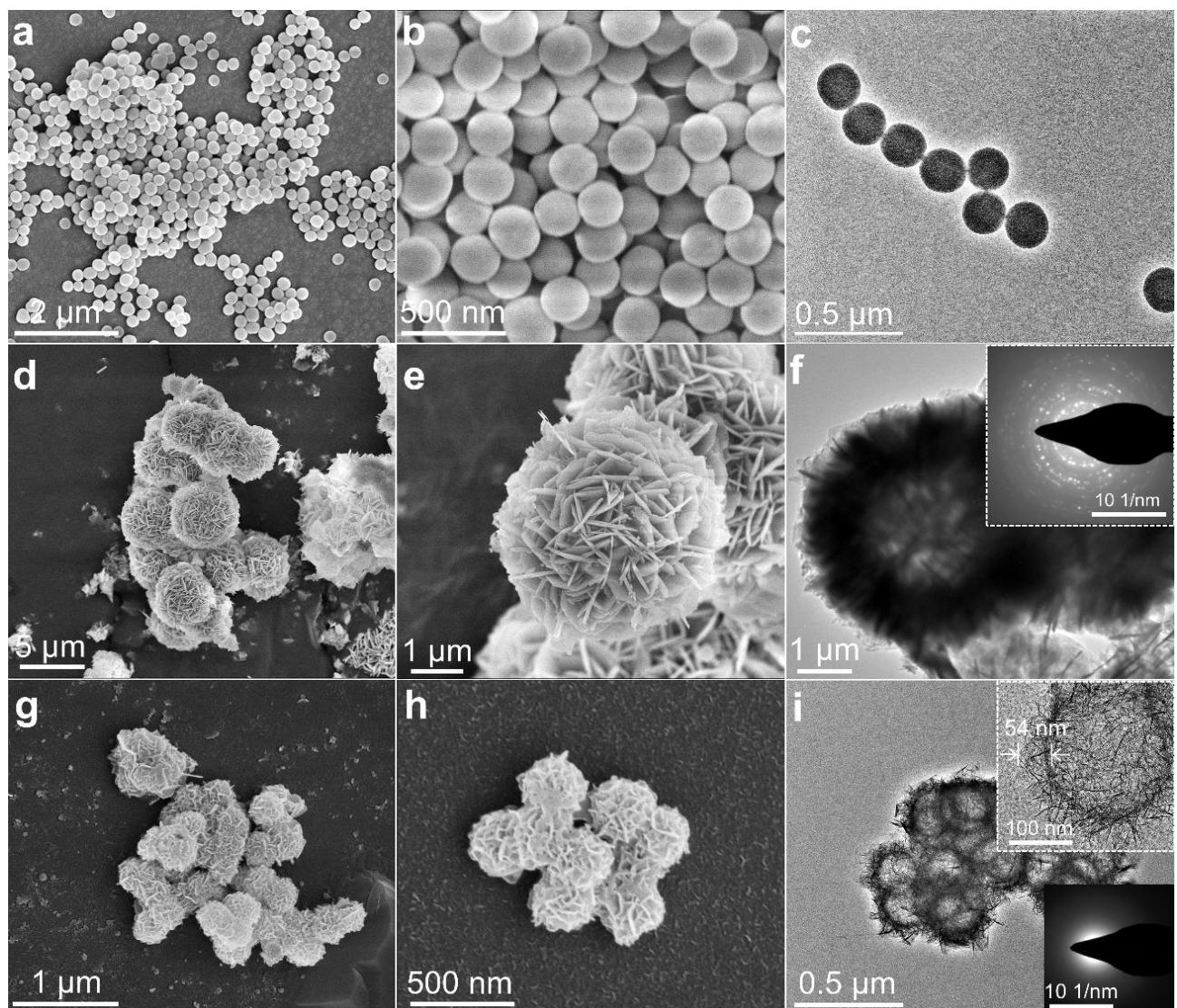


Figure S1. (a-b)(d-e)(g-h) FE SEM images of the SiO<sub>2</sub> sphere, Ni(OH)<sub>2</sub> and NiSiOx. (c, f, i) TEM images of SiO<sub>2</sub> sphere, Ni(OH)<sub>2</sub> and NiSiOx. The insets in (f) and (i) are the SAED patterns of Ni(OH)<sub>2</sub> and NiSiOx.

**Figure S2**

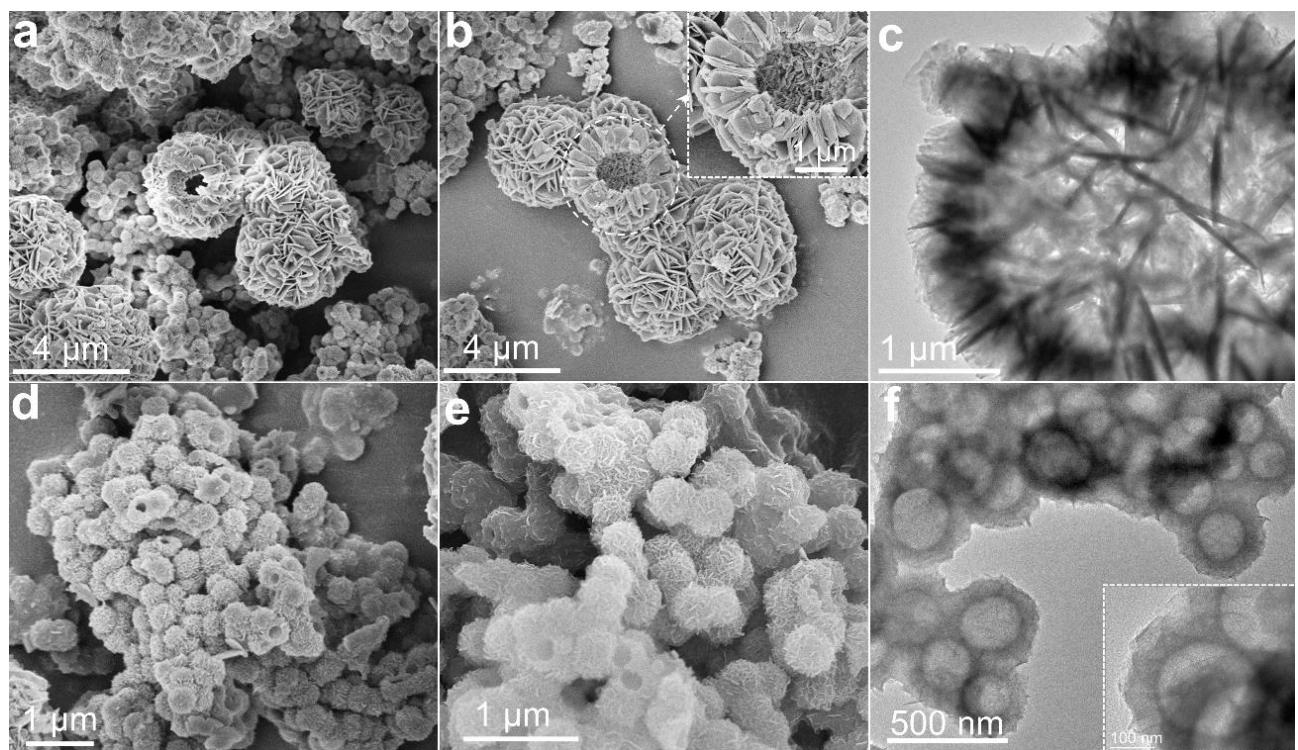


Figure S2. (a-b) FE SEM images of the  $\text{Ni(OH)}_2\text{-Si}$  in  $\text{NiSi-Ni(OH)}_2\text{-4}$ . (c)TEM image of the  $\text{Ni(OH)}_2\text{-Si}$  in  $\text{NiSi-Ni(OH)}_2\text{-4}$ . (d-e) FE SEM images of the  $\text{Ni(OH)}_2\text{@NiSi}$  in  $\text{NiSi-Ni(OH)}_2\text{-4}$ . (f) TEM image of the  $\text{Ni(OH)}_2\text{@NiSi}$  in  $\text{NiSi-Ni(OH)}_2\text{-4}$ .

**Figure S3**

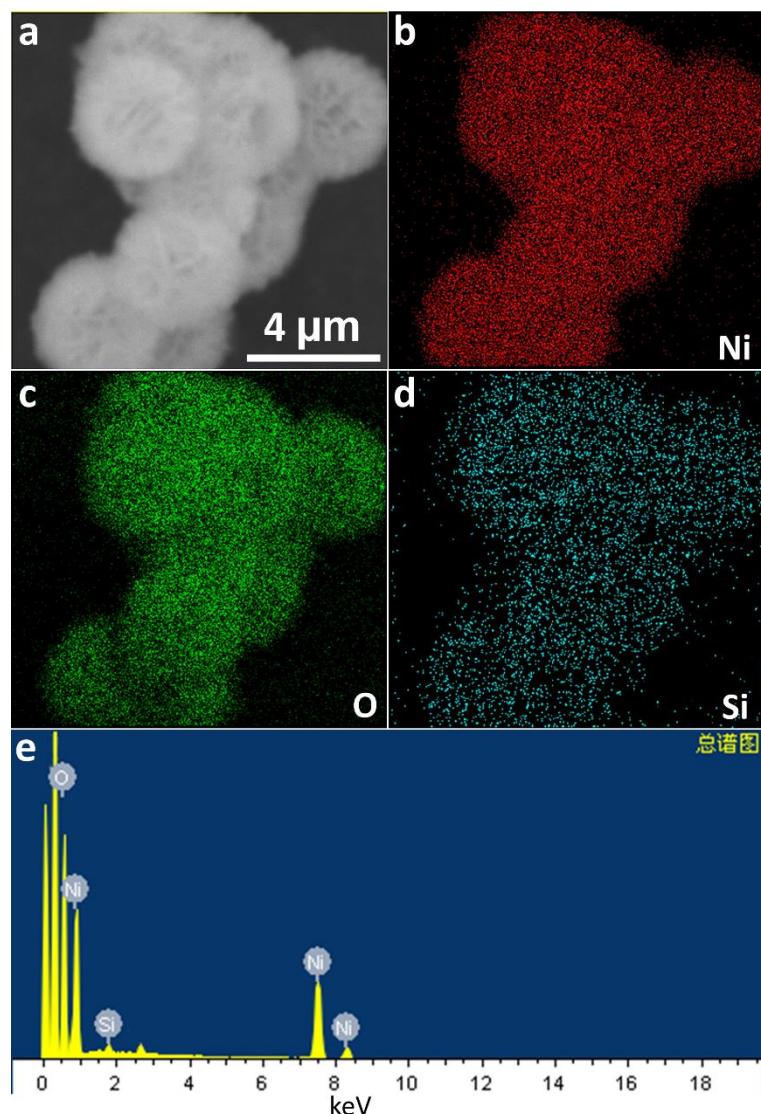


Figure S3. The selected SEM area (a), Elemental mapping images of (b) Ni, (c) O, (d) Si elements and EDX spectrum (e) of  $\text{Ni(OH)}_2$ -Si in  $\text{NiSi-Ni(OH)}_2$ -4.

**Figure S4**

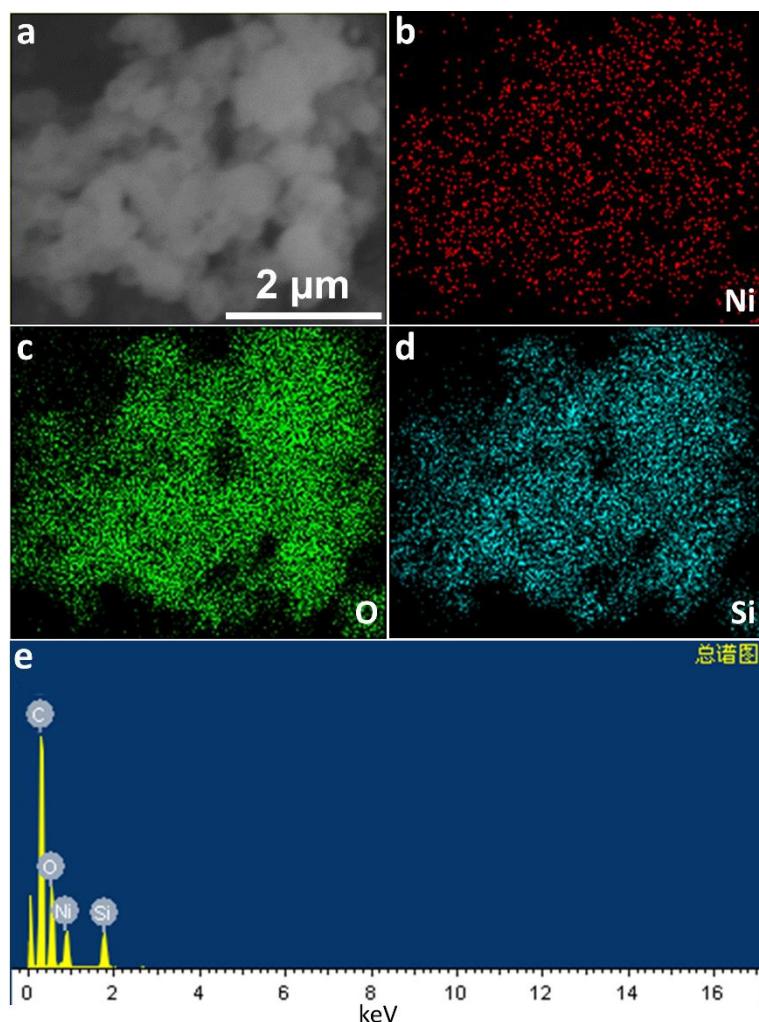


Figure S4. The selected SEM area (a), Elemental mapping images of (b) Ni, (c) O, (d) Si elements and EDX spectrum (e) of NiSi@Ni(OH)<sub>2</sub> in NiSi-Ni(OH)<sub>2</sub>-4.

**Figure S5**

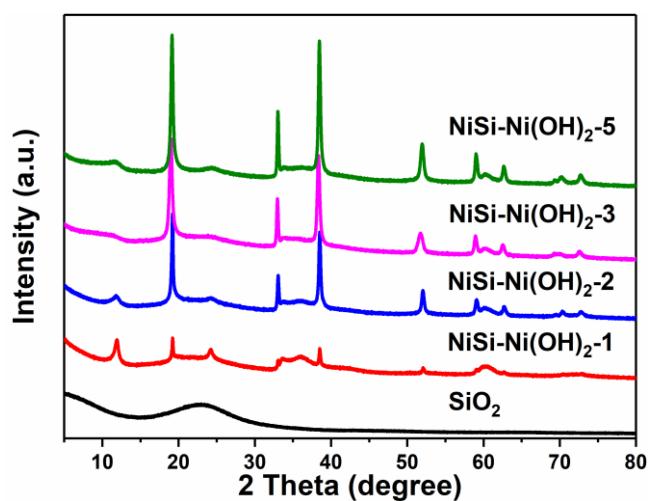


Figure S5. XRD patterns of  $\text{SiO}_2$ ,  $\text{NiSi-Ni(OH)}_2\text{-1}\sim\text{3}$  and  $\text{NiSi-Ni(OH)}_2\text{-5}$ .

**Figure S6**

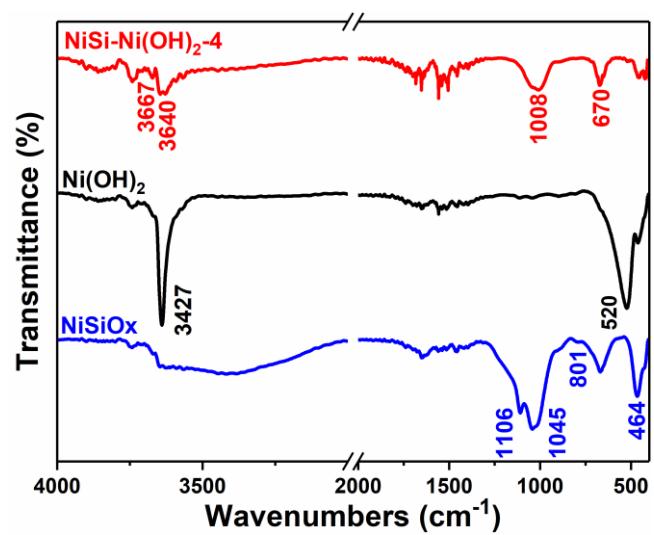


Figure S6. FT IR spectra of NiSiOx, Ni(OH)<sub>2</sub> and NiSi-Ni(OH)<sub>2</sub>-4.

**Figure S7**

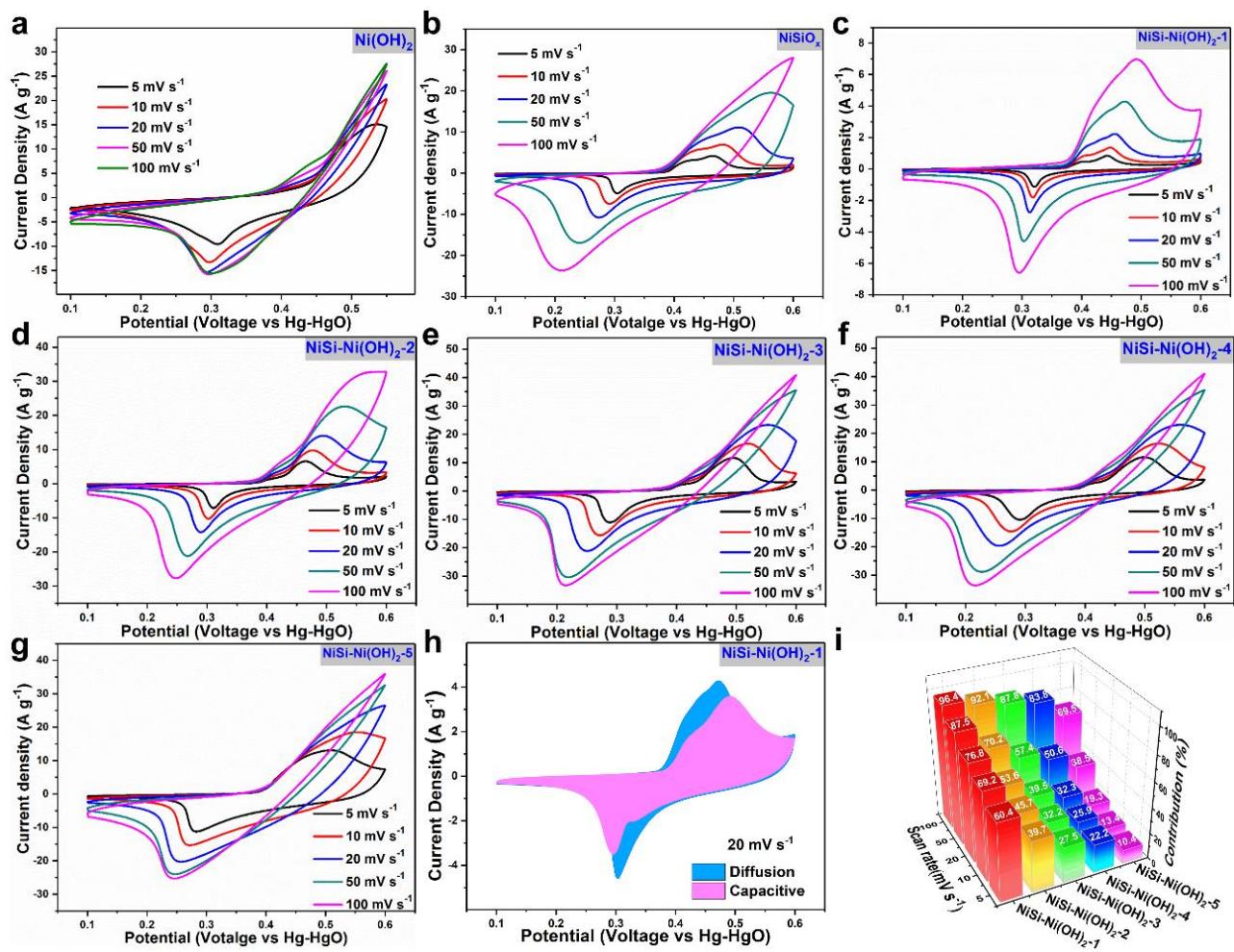


Figure S7. CV curves of (a)  $\text{Ni(OH)}_2$ , (b)  $\text{NiSiOx}$  and (c-g)  $\text{NiSi-Ni(OH)}_2-1\sim 5$  at different scan rates

( $5, 10, 20, 50$  and  $100 \text{ mV s}^{-1}$ ). (h) Capacitive and diffusion-controlled contributions at  $20 \text{ mV s}^{-1}$  of  $\text{NiSi-Ni(OH)}_2-1$ . (i) Capacitive contribution at different scan rates of  $\text{NiSi-Ni(OH)}_2-1\sim 5$ .

**Figure S8**

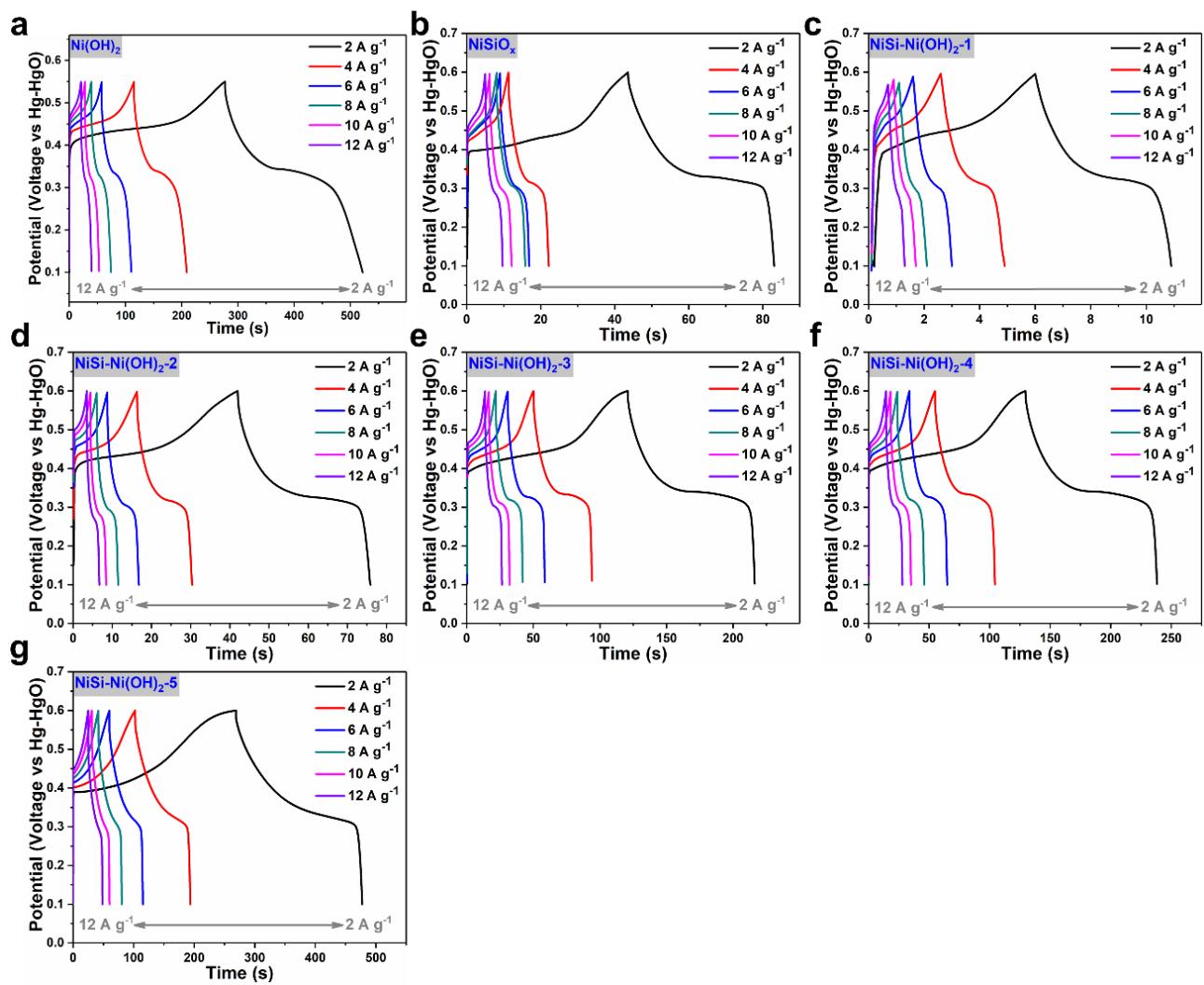


Figure S8. GCD curves of (a) Ni(OH)<sub>2</sub>, (b) NiSiO<sub>x</sub> and (c-h) NiSi-Ni(OH)<sub>2</sub>-1~5 at different discharge currents (2, 4, 6, 8, 10 and 12 A g<sup>-1</sup>).

**Figure S9**

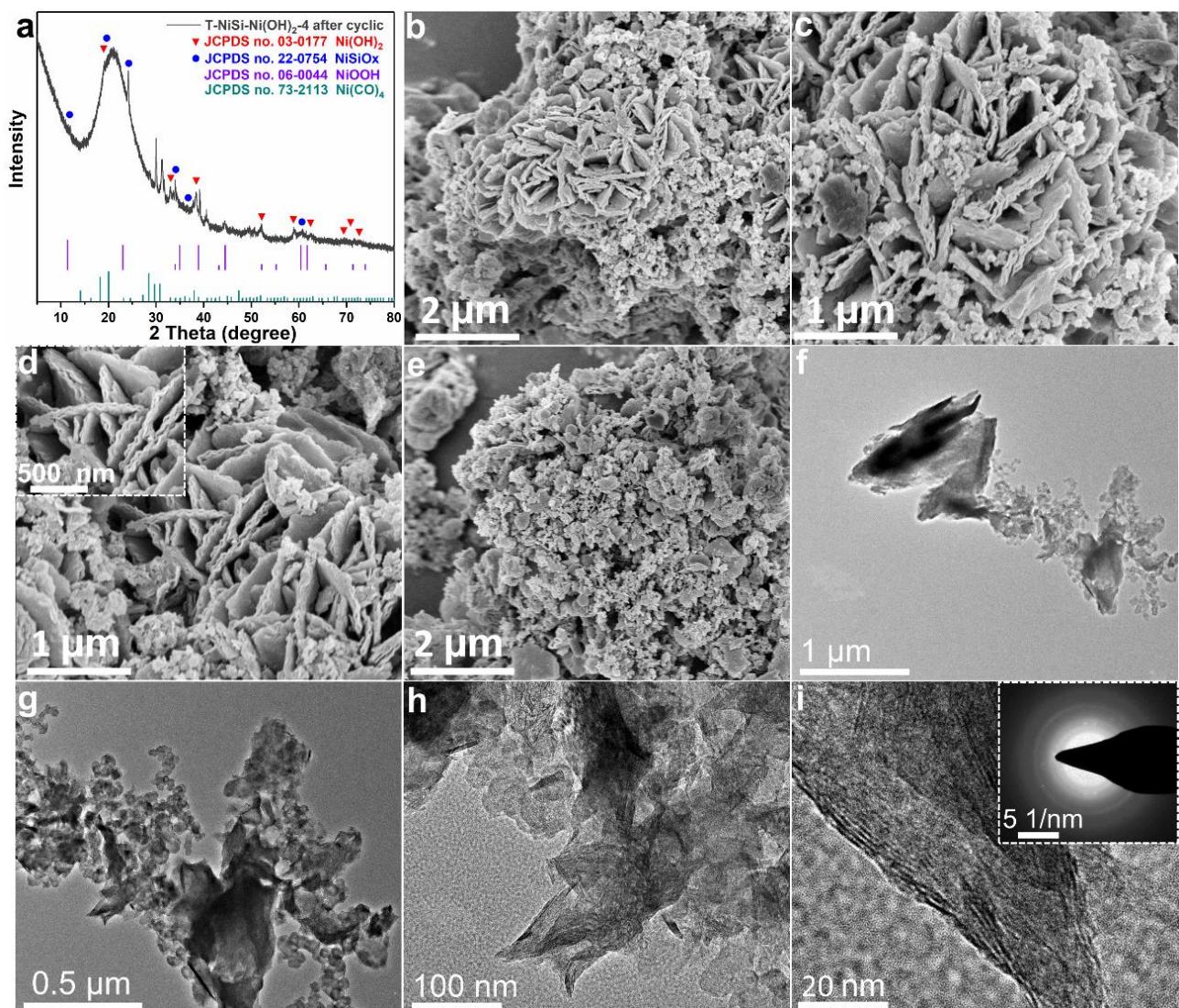


Figure S9. (a) XRD patterns of NiSi-Ni(OH)<sub>2</sub>-4 after cycling. (b-e) FE-SEM images of NiSi-Ni(OH)<sub>2</sub>-4 after cycling. (f-i) TEM and HR-TEM images of NiSi-Ni(OH)<sub>2</sub>-4 after cycling. (h-i) FE-SEM images of NiSi after cycling.

**Figure S10**

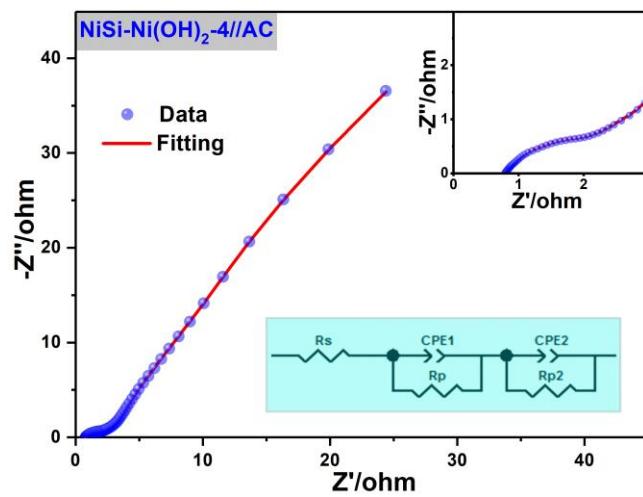


Figure S10. Nyquist plots of the HSC. The insets are the enlarged Nyquist plot from the high-frequency region and the equivalent circuit for the electrochemical impedance spectrum.

**Table S1****Table S1.** Comparison of the electrochemical performance with work from literatures based on nickel silicate/nickel hydroxide-based materials.

Nickel silicate/nickel hydroxide-based materials	Electrolyte	Potential /V	Capacitance	Cycle	Ref.
(Ni, Co) <sub>3</sub> Si <sub>2</sub> O <sub>5</sub> (OH) <sub>4</sub>	1 M KOH	0~0.5	144 F g <sup>-1</sup> , 1 A g <sup>-1</sup>	99.3%, 10000 cycles	<sup>1</sup>
Ni <sub>3</sub> Si <sub>2</sub> O <sub>5</sub> (OH) <sub>4</sub>	6 M KOH	0~0.5	887 F g <sup>-1</sup> , 0.7 A g <sup>-1</sup>	96.8%, 2000 cycles	<sup>2</sup>
Ni <sub>3</sub> Si <sub>2</sub> O <sub>5</sub> (OH) <sub>4</sub> /RGO	2 M KOH	0.2~0.6	178.9 F g <sup>-1</sup> , 1 A g <sup>-1</sup>	97.6%, 5000 cycles	<sup>1</sup>
C/Ni <sub>3</sub> Si <sub>2</sub> O <sub>5</sub> (OH) <sub>4</sub>	3 M KOH	-1~0.3	132.4 F·g <sup>-1</sup> , 0.5 A·g <sup>-1</sup>	100%, 10000 cycles	<sup>3</sup>
Ni <sub>3</sub> Si <sub>2</sub> O <sub>5</sub> (OH) <sub>4</sub> /rGO microspheres	2M KOH	0.15~0.65	178.9F·g <sup>-1</sup> , 1 A·g <sup>-1</sup>	97.6%,5000 cycles	<sup>1</sup>
NiSi hollow sphere	3 M KOH	0~0.6	66.7 F g <sup>-1</sup> , 0.5 A g <sup>-1</sup>	44%, 5000 cycles	<sup>4</sup>
NG/Ni(OH) <sub>2</sub>	2 M KOH	0~0.5	782 F g <sup>-1</sup> , 0.2 A g <sup>-1</sup>	90%, 10000 cycles	<sup>5</sup>
Nickel hydroxide	1 M KOH	0.1~0.6	3637 F g <sup>-1</sup> , 1 A g <sup>-1</sup>	80%, 10000 cycles	<sup>6</sup>
$\beta$ -Ni(OH) <sub>2</sub>	6 M KOH	0.1~0.55	2537.4 F g <sup>-1</sup> , 1 A g <sup>-1</sup>	77.6%, 3000 cycles	<sup>7</sup>
Ni(OH) <sub>2</sub> nanosheets	2 M KOH	0~0.45	2384.3 F g <sup>-1</sup> , 1 A g <sup>-1</sup>	75%, 3000 cycles	<sup>8</sup>
Ni(OH) <sub>2</sub> -PPy	3 M KOH	0~0.5	352 F g <sup>-1</sup> , 1 A g <sup>-1</sup>	87%, 7000 cycles	<sup>9</sup>
Ni(OH) <sub>2</sub> -POV	2 M KOH	0~0.45	1440 F g <sup>-1</sup> , 1 A g <sup>-1</sup>	85%, 2000 cycles	<sup>10</sup>
Ni(OH) <sub>2</sub> nanotubes	6 M KOH	-0.15~0.45	1319 F g <sup>-1</sup> , 3 A g <sup>-1</sup>	79.3%, 7000 cycles	<sup>11</sup>
Nickel hydroxide–nickel	3 M KOH	0~0.5	1793 F g <sup>-1</sup> , 1.25 A g <sup>-1</sup>	97.2%, 3000 cycles	<sup>12</sup>
Nickel hydroxide	1 M KOH	0~0.5	2801 F g <sup>-1</sup> , 2 A g <sup>-1</sup>	83%, 1500 cycles	<sup>13</sup>
HCNs@NiCo-LDH	6 M KOH	0~0.6	2558 F g <sup>-1</sup> , 1 A g <sup>-1</sup>	77%, 10000 cycles	<sup>14</sup>
Ag NW/Ni(OH) <sub>2</sub>	1 M KOH	0~0.6	164.8 F g <sup>-1</sup> , 5 mV s <sup>-1</sup>	62.5%, 1000 cycles	<sup>15</sup>
Ni <sub>1</sub> Co <sub>2</sub>	3 M KOH	0~0.45	2654.9 F g <sup>-1</sup> , 1 A g <sup>-1</sup>	77%, 1500 cycles	<sup>16</sup>
CNT@Ni(OH) <sub>2</sub>	1 M KOH	0~0.5 V	1136 F g <sup>-1</sup> at 2 A g <sup>-1</sup>	92%, 1000 cycles	<sup>17</sup>
NiSi-Ni(OH) <sub>2</sub>	3M KOH	0.1~0.6	476.4 F·g <sup>-1</sup> , 2 A·g <sup>-1</sup>	103.3%, 10000 cycles	This work

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