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## Supplementary material Hierarchical flower-like Ni-Co layered double hydroxide nanostructures: synthesis and super performance

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Fig. S1 (a) SEM image and (b) XRD pattern of Ni(OH)<sub>2</sub>.



Fig. S2 (a) SEM image and (b) XRD pattern of Co(OH)<sub>2</sub>.



Fig. S3 EDS analysisof (a) Ni-Co LDH 1/2, (b) Ni-Co LDH 1/1 and (c) Ni-Co LDH 2/1.

| r Atomic percentage | es of NI, Co, and O el        | lements.  |
|---------------------|-------------------------------|---|
| Ni                  | Со                            | 0   |
| 11.87               | 23.95                         | 64.18   |
| 14.90               | 15.68                         | 69.42   |
| 24.78               | 12.41                         | 62.81   |
|                     | Ni<br>11.87<br>14.90<br>24.78 | Ni Co   11.87 23.95   14.90 15.68   24.78 12.41 |

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Fig. S4 (a) SEM image and (b) XRD pattern of product prepared with H<sub>2</sub>O as the solvent.



Fig. S5 XRD pattern of Ni(OH)<sub>2</sub> prepared with H<sub>2</sub>O as the solvent.



Fig. S6 Raman spectra of Ni-Co LDH 1/2, Ni-Co LDH 1/1, Ni-Co LDH 2/1 and ethanol.



Fig. S7 XPS spectra of Ni-Co LDH 1/2, Ni-Co LDH 1/1 and Ni-Co LDH 2/1 for full survey scan.

|                     | Pseudo first-o      | Pseudo second-order models           |                  |                |                                      |                  |                |
|---------------------|---------------------|--------------------------------------|------------------|----------------|--------------------------------------|------------------|----------------|
| Sample              | $q_{e} (mg g^{-1})$ | q <sub>e</sub> (mg g <sup>-1</sup> ) | ) K <sub>1</sub> | R <sup>2</sup> | q <sub>e</sub> (mg g <sup>-1</sup> ) | ) K <sub>2</sub> | R <sup>2</sup> |
|                     | experiment          | model                                |                  |                | model                                |                  |                |
| Ni-Co LDH 1/2       | 230.5               | 243.3                                | 1.04             | 0.92698        | 253.8                                | 0.00303          | 0.99779        |
| Ni-Co LDH 1/1       | 252.6               | 291.5                                | 0.25             | 0.95621        | 275.5                                | 0.0188           | 0.99772        |
| Ni-Co LDH 2/1       | 211.1               | 231.5                                | 1.81             | 0.99405        | 234.8                                | 0.00223          | 0.99966        |
| Ni(OH) <sub>2</sub> | 229.2               | 241.3                                | 2.13             | 0.95499        | 236.4                                | 0.00144          | 0.99275        |
| Co(OH) <sub>2</sub> | 224.6               | 251.2                                | 1.79             | 0.98102        | 243.9                                | 0.00268          | 0.99926        |

Table S2. Kinetics models fitted data of AF adsorption with different adsorbents.

Table S3. Kinetics models fitted data of CR adsorption with different adsorbents.

|                     | Pseudo first-order models                          |   |                |                | Pseudo second-order models                    |                      |                |
|---------------------|--|---|----------------|----------------|---|----------------------|----------------|
| Sample              | q <sub>e</sub> (mg g <sup>-1</sup> )<br>experiment | q <sub>e</sub> (mg g <sup>-1</sup> )<br>model | K <sub>1</sub> | R <sup>2</sup> | q <sub>e</sub> (mg g <sup>-1</sup> )<br>model | ) K <sub>2</sub>     | R <sup>2</sup> |
| NE CELDII 1/2       | 172.4  | 102.0   | 2.00           | 0.07529        | 104.0   | 1.2*10-2             | 0.00701        |
| N1-C0 LDH 1/2       | 1/3.4  | 183.8   | 3.86           | 0.9/538        | 184.8   | 1.3*10-2             | 0.99/91        |
| Ni-Co LDH 1/1       | 245.2  | 259.7   | 0.59           | 0.97401        | 261.1   | 6.1*10-2             | 0.99839        |
| Ni-Co LDH 2/1       | 148.5  | 184.5   | 8.24           | 0.97171        | 193.4   | 5.1*10-3             | 0.99703        |
| Ni(OH) <sub>2</sub> | 165.8  | 159.7   | 7.18           | 0.91864        | 172.4   | 5.8*10 <sup>-3</sup> | 0.99291        |
| Co(OH) <sub>2</sub> | 163.4  | 174.2   | 4.33           | 0.96964        | 174.8   | 7.9*10 <sup>-3</sup> | 0.99987        |

Table S4. Adsorption models fitted data of AF and CR adsorption onto Ni-Co LDH 1/1.

|            | Langmuir 1            | nodels                |                |                | Freundlic      | h models | 5              |
|------------|-----------------------|-----------------------|----------------|----------------|----------------|----------|----------------|
| Adsorption | K <sub>L</sub>        | q <sub>max</sub>      | R <sub>L</sub> | R <sup>2</sup> | K <sub>F</sub> | n        | R <sup>2</sup> |
|            | (L mg <sup>-1</sup> ) | (mg g <sup>-1</sup> ) |                |                | $(L mg^{-1})$  |          |                |
| AF         | 0.037                 | 2787                  | 0.043-0.119    | 0.99486        | 812.4          | 5.01     | 0.88851        |
| CR         | 0.0424                | 1915.7                | 0.038-0.106    | 0.99690        | 937.7          | 9.69     | 0.90211        |



Fig. S8 Effect of dye concentrations on the equilibrium adsorption capacity of AF and CR adsorption onto Ni-Co LDH 1/1.



Fig. S9 (a) Zeta potential of Ni-Co LDH 1/1 at different pH values. (b) Zeta potential of Ni-Co LDH 1/1, Ni-Co LDH 1/2, Ni-Co LDH 2/1, Ni(OH)<sub>2</sub> and Co(OH)<sub>2</sub> at pH values of 7.

| Adsorbents                                     | $q_{max} (mg g^{-1})$ | References |
|--|-----------------------|------------|
|  | 2787(AF)              | This moult |
| NI-COLDH 1/1                                   | 1915.7 (CR)           | THIS WOLK  |
| C/NiFe <sub>2</sub> O <sub>4</sub>             | 21(AF)                | [1]        |
| NiOnanosheets                                  | 22(AF)                | [2]        |
| Chitosan                                       | 43(AF)                | [3]        |
| Graphene oxide/chitosan                        | 130(AF)               | [4]        |
| Montmorillonite                                | 161(AF)               | [5]        |
| Carbon-alumina composite                       | 95 (AF)               | [6]        |
| ZnFe <sub>2</sub> O <sub>4</sub> hollow fibers | 150(AF)               | [7]        |
| LTA-type zeolite                               | 40(AF)                | [8]        |
| CLDH/y-AlO(OH)-2-500                           | 447 (CR)              | [9]        |
| MgO (111) nanoplates                           | 303 (CR)              | [10]       |
| Activated carbon powder                        | 500 (CR)              | [11]       |
| CoFe <sub>2</sub> O <sub>4</sub>               | 244.5 (CR)            | [12]       |
| Activated carbon from coal                     | 189 (CR)              | [13]       |

Table S5 Comparison of maximum adsorption capacitance for acid fuchsin of various adsorbents.

Table S6 Comparisons of the specific capacitances for Ni-Co LDHs based electrode materials in three-electrode system.

| Ni-Co LDHs based electrodes                               | Specific capacitances(F g <sup>-1</sup> ) | Ref. |
|---|---|------|
|   | 21(0, 2, (1, A, -1))                      | This |
| NI-CO LDH 1/2   | $3108.3(1 \text{ A g}^{-1})$              | work |
| Ni-Co LDHs on conducting Zn <sub>2</sub> SnO <sub>4</sub> | 1805 (0.5 A g <sup>-1</sup> )             | [14] |
| Nickel/cobalt double hydroxides                           | 1887.5 (1 A g <sup>-1</sup> )             | [15] |
| Ni-Co LDH nanosheets                                      | 2682 (3 A g <sup>-1</sup> )               | [16] |
| Vertically aligned Ni-Co LDHs nanosheet                   | 1734 (6 A g <sup>-1</sup> )               | [17] |
| Ni-Co LDHs nanoflakes/carbon cloth                        | 1938 (1 A g <sup>-1</sup> )               | [18] |
| Carbon nanotubes/Ni-Co hydroxide nanoflake                | 1151 (1 A g <sup>-1</sup> )               | [19] |
| Flower-like nickel-cobalt binary hydroxides               | 1804 (1 A g <sup>-1</sup> )               | [20] |
| Nitrogen-Doped Carbon Nanofiber/Ni-Co LDHs                | 1950 (1 A g <sup>-1</sup> )               | [21] |



Fig. S10 XRD patterns of Ni-Co LDH 1/1 before and after AF and CR adsorption.



Fig. S11 CV and galvanostatic charge/discharge curves: (a) and (b) Ni(OH)<sub>2</sub> electrode, (c) and (d) Co(OH)<sub>2</sub> electrode, (e) and (f) Ni-Co LDH 1/1electrode and (g) and (h) Ni-Co LDH 2/1 electrode.



Fig. S12 (a) Galvanostatic charge/discharge curves of Ni-Co LDH with molar ratio of Ni/Co is 1:4, inset shows the SEM image. (b) Relationship plot between discharge specific capacitance and molar ratio of Ni/Co at current density of 1 A g<sup>-1</sup>.



Fig. S13 SEM image of Ni-Co LDH 1/2 electrode after 3000 cycles.

The specific capacitances (SC) were calculated using the integral current areas of galvanostatic discharge curve with following formula <sup>22</sup>:

$$SC = \frac{2I_m \int Vdt}{V_i^2 - V_f^2}$$

 $I_m$  (A g<sup>-1</sup>) is the current density, where I is the current and m is the mass of active material.  $\int V dt$ 

is the integral current area, where V is the potential with initial and final values of  $V_i$  and  $V_f$ , respectively. In this work, the area of the working electrode immersed into the electrolyte was controlled to be about 1 cm<sup>2</sup>.

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