Supplementary Information

Fabrication of hierarchically porous monolithic layered double hydroxide composites with tunable microcages for effective oxyanion adsorption
Naoki Tarutani a, Yasuaki Tokudome a*, Megu Fukui, a Kazuki Nakanishi b, and Masahide Takahashi a

\[ a \text{ Department of Materials Science, Graduate School of Engineering, Osaka Prefecture University, Sakai, Osaka 599-8531, Japan} \]
\[ b \text{ Department of Chemistry, Graduate School of Science, Kyoto University, Kitashirakawa, Sakyo-ku, Kyoto 606-8502, Japan} \]

Supplementary Figures

- Figure S1: 2
- Figure S2: 3
- Figure S3: 4
- Figure S4: 5
- Figure S5: 6
Figure S1. Schematic illustration of LDH crystal. Upper image shows microcage size, $d_{\text{micro}}$, which is calculated from hydroxide sheet thickness, $t_{\text{sheet}}$, and (003) lattice spacing, $d_{003}$, as follows: $d_{\text{micro}} = t_{\text{sheet}} + (d_{003} - t_{\text{sheet}})/2$. $t_{\text{sheet}}$ is 200 pm$^1$ and $d_{003}$ is estimated from the XRD pattern. The lower image shows adsorption of an oxyanion, in which the oxyanion fits into a microcage formed between adjacent octahedral metal hydroxides.

Reference
Figure S2. Magnified SEM image of Figure 1 (b).
Figure S3. XRD patterns of Mg-Al type LDH monolithic composites prepared with various Mg/Al molar ratios from 0.1 to 3.0.
Figure S4. Time evolutions of solution pH after PO addition in Mg-Al system. In the curves, pH rapidly increased to about 3.0 within several minutes (1st stage), showed a plateau (2nd stage, formation of Al(OH)$_3$), rapidly increased again (3rd stage), and then showed a gradual increase (4th stage, formation of LDH and/or Mg(OH)$_2$).
Figure S5. $\text{CrO}_4^{2-}$ adsorption amounts, specific surface areas (red circle) and crystallite sizes (green square) of M(II)-Al type LDH composites (M(II) = Mg, Mn, Fe, Co, and Ni).