

## **Electronic Supplementary Information**

### **Precise size control of layered double hydroxide nanoparticles through reconstruction using tripodal ligands**

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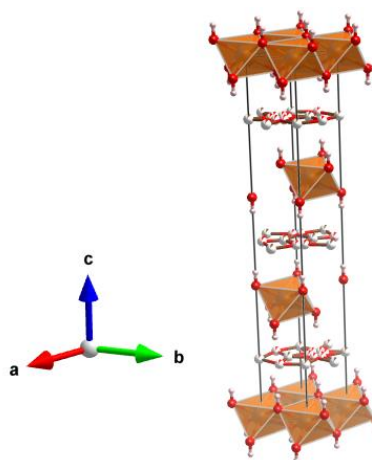
### Theoretical Surface Area of an LDH sheet

Consider about an LDH sheet with unit cell size in lateral directions and the composition of  $\text{Mg}_{0.667}\text{Al}_{0.333}(\text{OH})_2 \cdot 0.167\text{CO}_3$ . Interlayer water is ignored because BET surface area is measured for its dried state. The surface area on both top and bottom surfaces of a unit-cell-size sheet is  $\sqrt{3}a^2 = 0.161 \times 10^{-18} \text{ m}^2$ , where  $a = 0.305 \text{ nm}$ . The mass of a unit-cell-size sheet is

$$\begin{aligned} & \{24.31 \times 0.667 + 26.98 \times 0.333 + 16.00 \times (2 + 0.167 \\ & \times 3) + 1.008 \times 2 + 12.01 \times 0.167\} / (6.02 \times 10^{23}) \\ & = 11.50 \times 10^{-23} \text{ g.} \end{aligned}$$

The theoretical surface area is

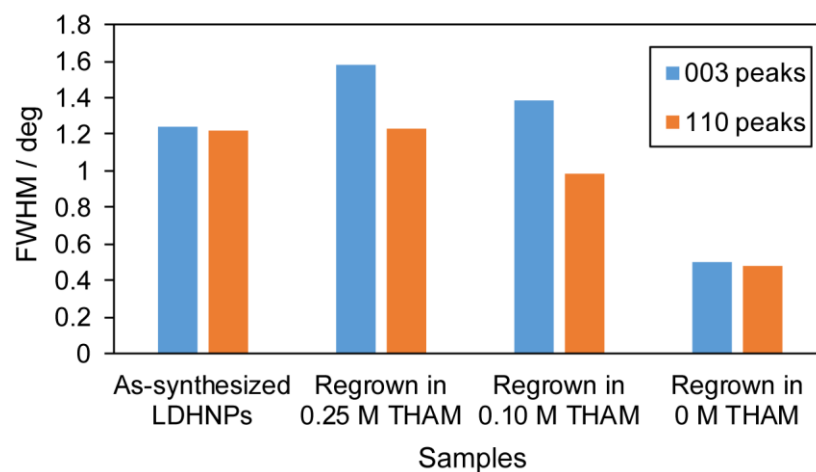
$$(0.161 \times 10^{-18}) / (11.50 \times 10^{-23}) = 1400 \text{ m}^2/\text{g}$$



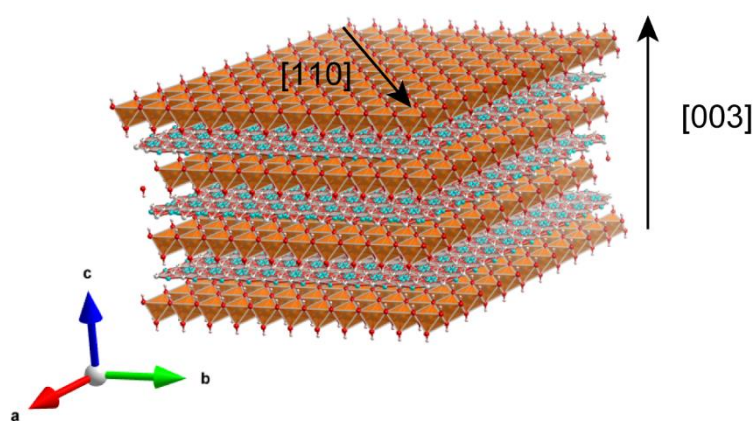
Unit cell image of an LDH.

**Table S1.** FWHM of diffraction peaks of LDHNPs regrown in dilute THAM solutions.

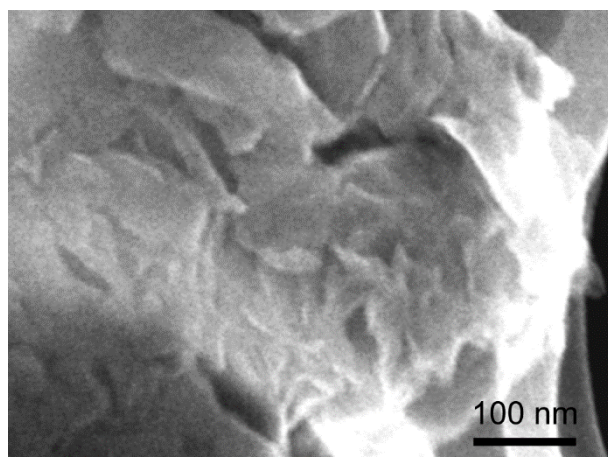
[THAM]/M	FWHM of 003 peaks/deg	FWHM of 110 peaks/deg
as-synthesized	1.24	1.22
0.25	1.58	1.22
0.10	1.39	0.98
0	0.50	0.48



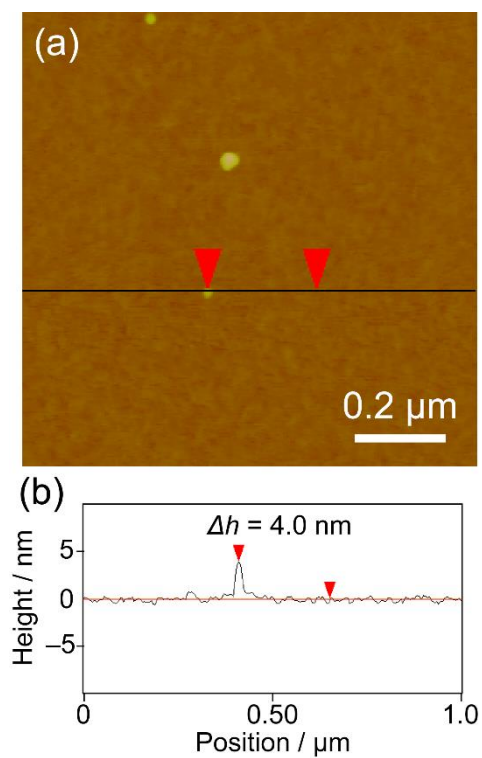
**Fig. S1.** FWHM of the diffraction peaks of LDHNPs regrown in dilute THAM solutions.



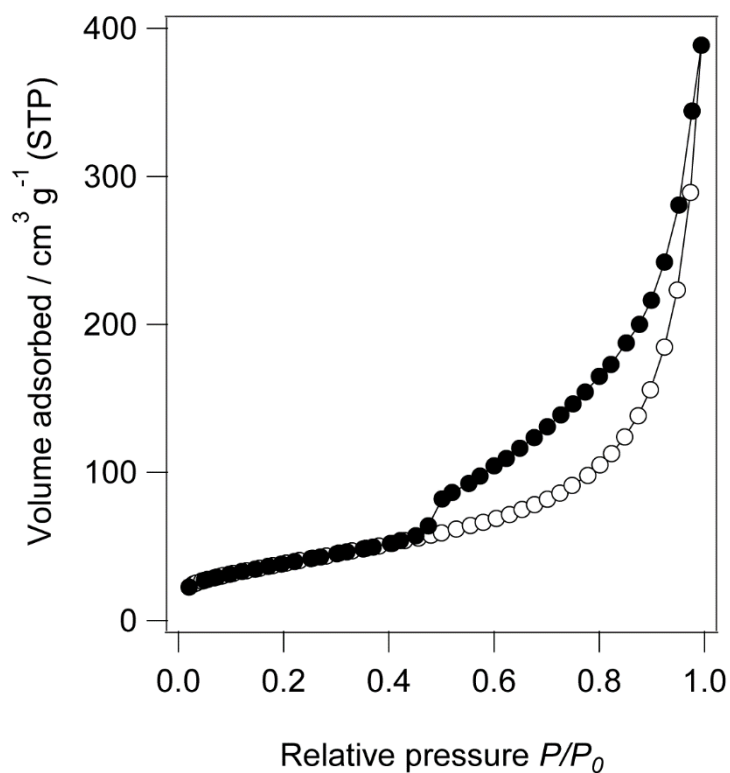
**Fig. S2.** Structural model of an LDH.



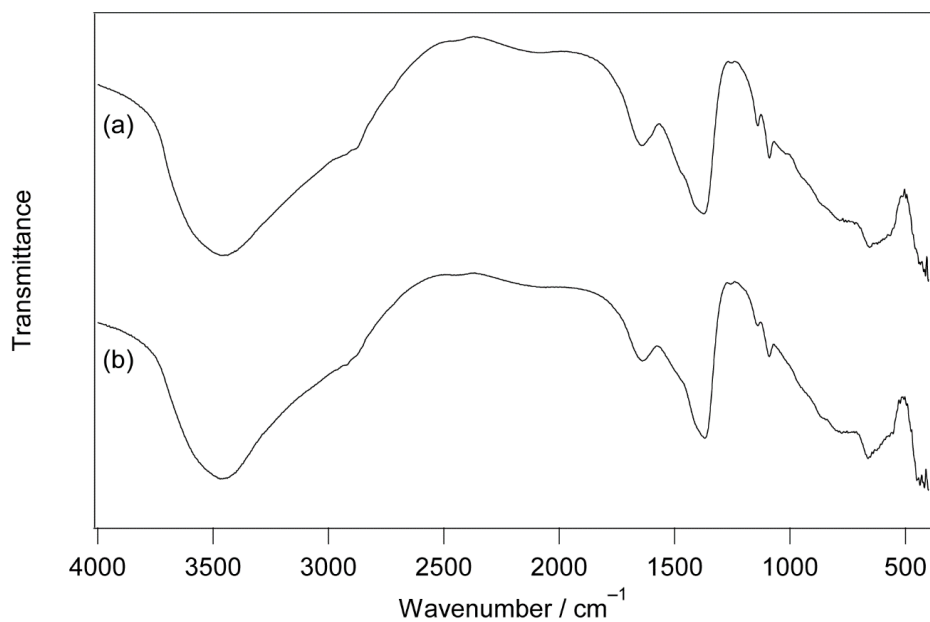
**Fig. S3.** SEM image of RLDH-1.0 without an ultrasonic treatment.



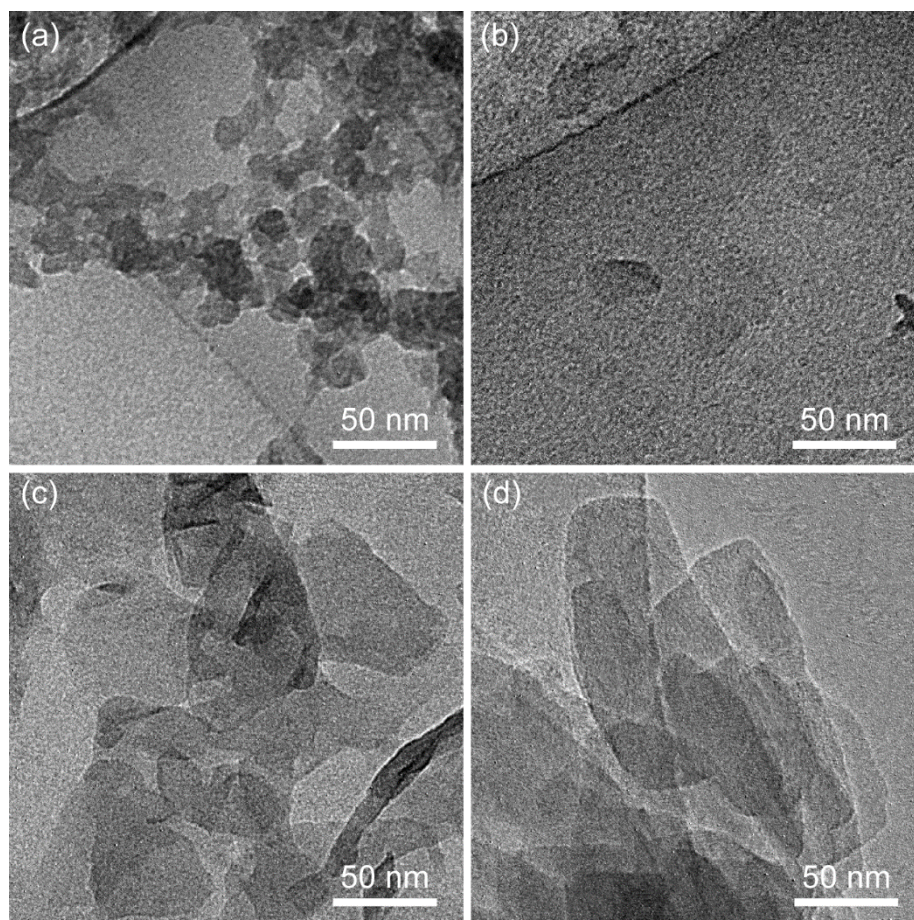
**Fig. S4.** (a) AFM image of RLDH-1.0. (b) The height profile on the line indicated in (a).



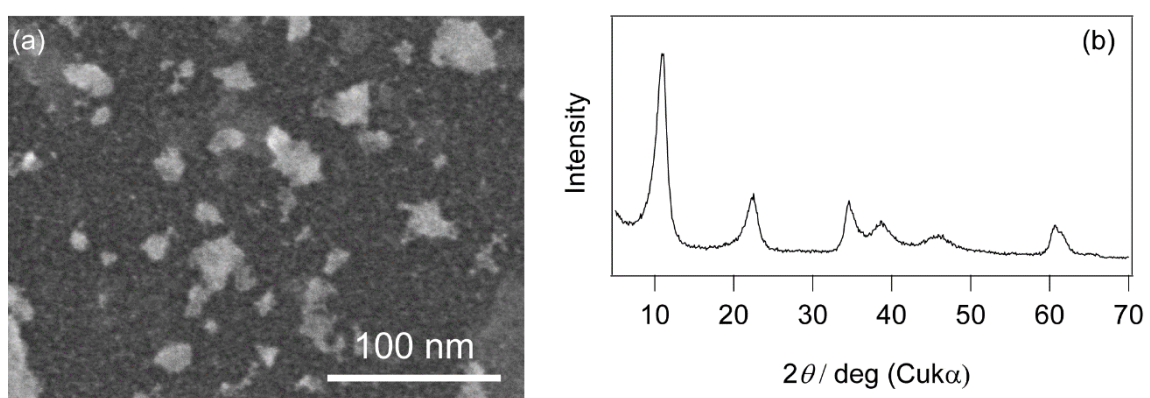
**Fig. S5.** N<sub>2</sub> adsorption–desorption isotherm of RLDH-1.0 without ultrasonic treatment.



**Fig. S6.** FTIR spectra of (a) LDHNPs synthesized by the modified coprecipitation, using THAM and (b) those synthesized by the reconstruction method using THAM. No significant differences were observed between these spectra.



**Fig. S7.** TEM images of (a) RLDH-1.0, (b) RLDH-0.50, (c) RLDH-0.20, and (d) RLDH-0.

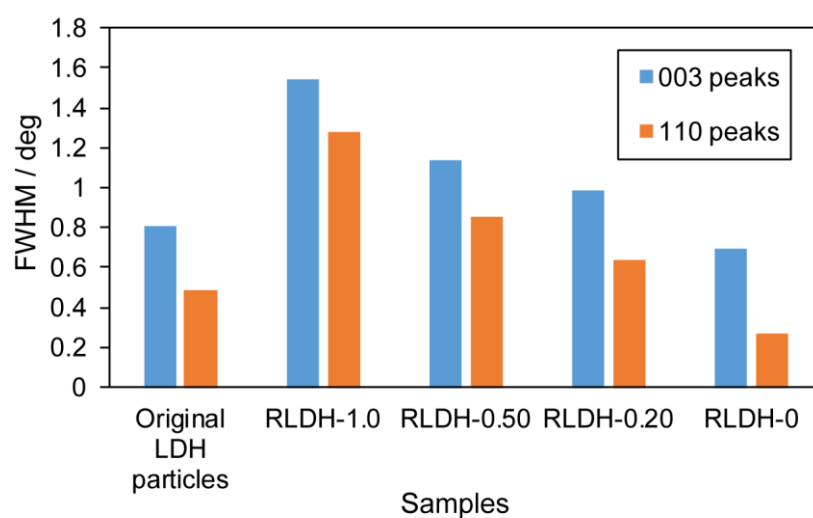


**Fig. S8.** (a) SEM image and (b) XRD pattern of LDHNPs reconstructed from MMO(85)-c by using pentaerythritol as a tripodal ligand.

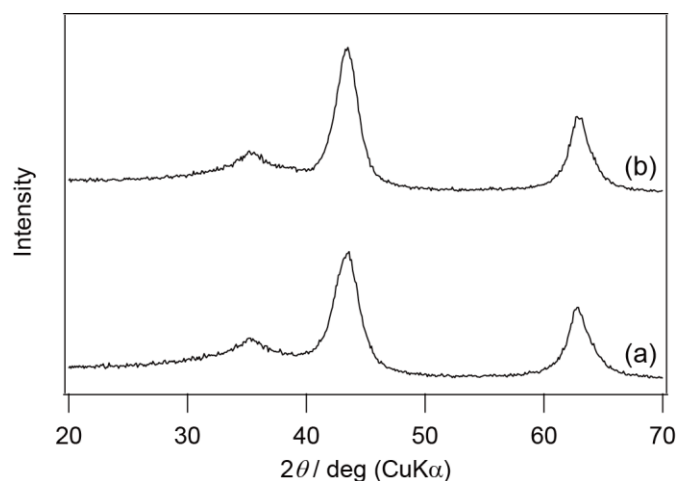


**Table S2.** FWHM of diffraction peaks of LDHNPs prepared by the reconstruction method.

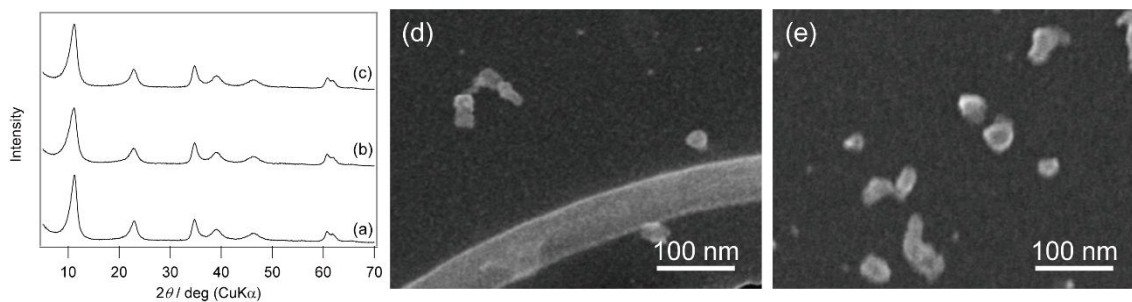
Sample	FWHM of 003 peaks/deg	FWHM of 110 peaks/deg
Original LDH particles	0.80	0.49
RLDH-0	0.70	0.27
RLDH-1.0	1.54	1.28
RLDH-0.50	1.14	0.86
RLDH-0.20	0.99	0.64



**Fig. S9.** FWHM of the diffraction peaks of LDHNPs prepared by the reconstruction method.



**Fig. S10.** XRD patterns of MMOs calcined at (a) 450 °C and (b) 550 °C.



**Fig. S11.** XRD patterns of RLDH-1.0 prepared (a) from MMO calcined at 450 °C without stirring during reconstruction, (b) from MMO calcined at 450 °C with stirring, and (c) from MMO calcined at 550 °C without stirring. SEM images of RLDH-1.0 prepared (d) from MMO calcined at 450 °C with stirring, and (e) from MMO calcined at 550 °C without stirring.