

Supporting Information for:

Controlling the Surface Morphology of Two Dimensional Nanomaterials upon Molecule-Mediated Crystal Growth

Tetsuo Yamaguchi,¹ Hyoung-Jun Kim,² Hee Jung Park,³ Taeho Kim,¹ Zubair Khalid,¹ Jin Kuen Park,^{4*} Jae-Min Oh^{1*}

¹Department of Energy and Materials Engineering, Dongguk University-Seoul, Seoul 04620, Korea

E-mail: jaemin.oh@dongguk.edu

²Research Institute, National Cancer Center, 323, Ilsan-ro, Goyang, Gyeonggi 10408, Republic of Korea

³ KBSI Western Seoul Center, University-Industry Cooperation Building, 150 Bugahyeon-ro, Seodaemun-gu, Seoul 03759, Republic of Korea

⁴ Department of Chemistry, Hankuk University of Foreign Studies, Yongin, 17035, Gyeonggi-do, Republic of Korea.

E-mail: jinkpark@hufs.ac.kr

- 1. Experimental**
- 2. AFM images**

1. Experimental

Table S1. Changes of pH values of methyl orange (MO) aqueous suspensions during the reconstruction of the LDHs.

Concentration (eq)	0.2	0.5	3.0
0 h (before adding LDO)	8.47	8.48	7.93
0.5 h	11.10	10.65	11.92
4 h	11.51	11.61	11.96
12 h	11.56	11.75	11.87

Table S2. Particle sizes (nm) of the reconstructed LDH₂₀₀₀ in CO₃²⁻ estimated by Sherrer's equation.

	0.2 eq	3 eq
0.5 h	5.40 nm	13.5 nm
4 h	4.66 nm	19.3 nm
12 h	8.63 nm	9.01 nm

Table S3. Particle sizes (nm) of the reconstructed LDH₃₅₀ in CO₃²⁻ estimated by Sherrer's equation.

	0.2 eq	3 eq
0.5 h	8.02 nm	9.03 nm
4 h	9.03 nm	11.4 nm
12 h	12.9 nm	13.7 nm

2. AFM images

Fig. S3 showed the 3-dimensional AFM images and corresponding height profile of pristine LDH and LDO. As clearly seen, the surface of both LDH and LDO, regardless of lateral dimension, was very flat (smooth). The surface roughness value Ra of LDH₂₀₀₀, LDO₂₀₀₀, LDH₃₅₀, and LDO₃₅₀ are 37, 35, 0.8 and 0.8 nm, respectively.

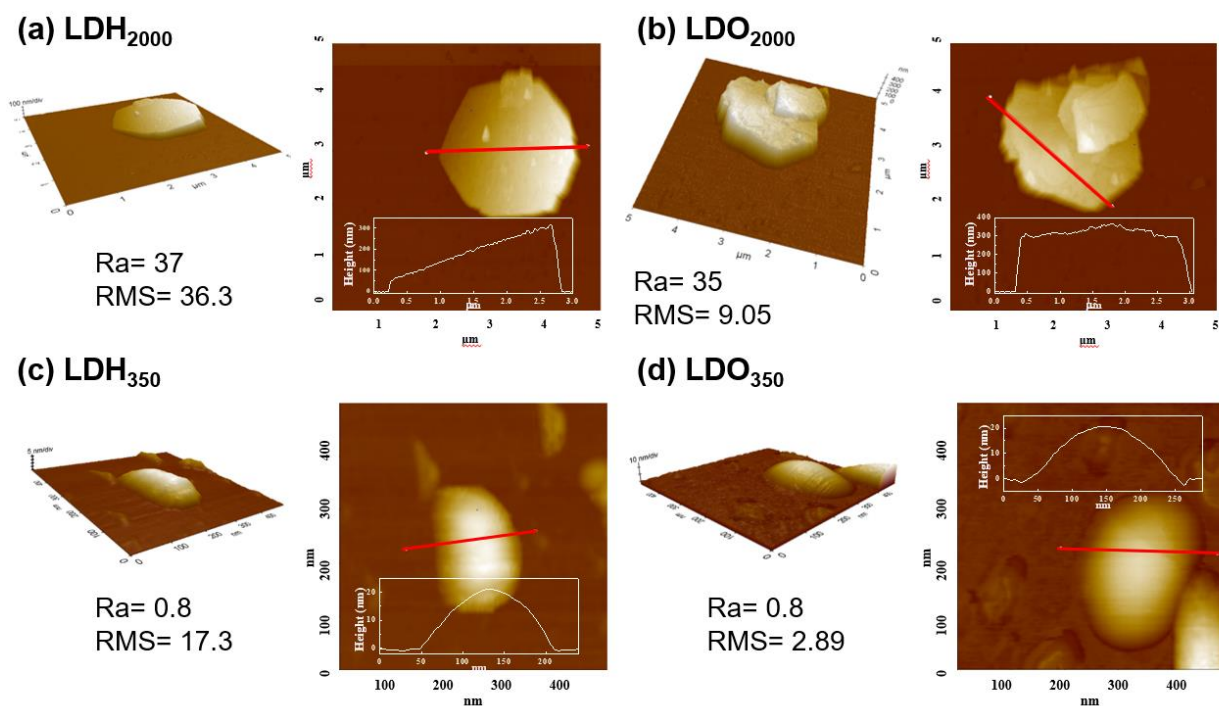


Fig. S1 Atomic force microscopic (AFM) images of (a) LDH₂₀₀₀, (b) LDO₂₀₀₀, (c) LDH₃₅₀ and (d) LDO₃₅₀.

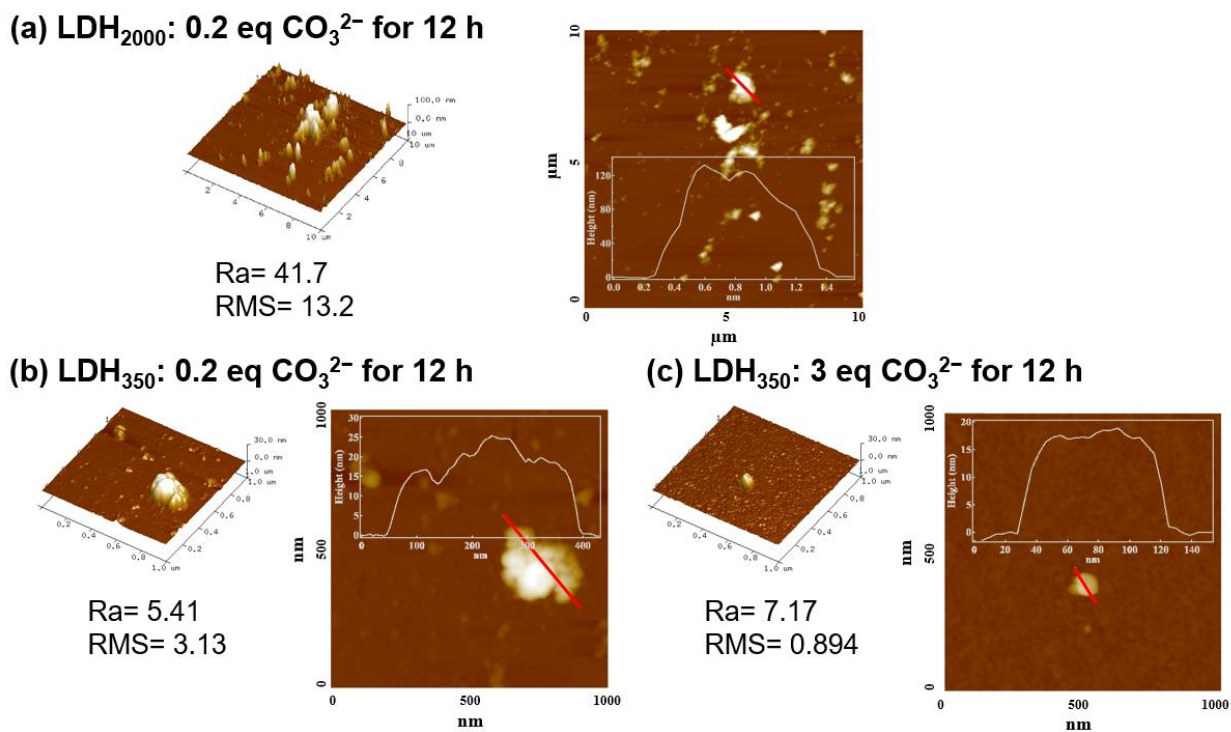


Fig. S2 Atomic force microscopic (AFM) images of the reconstructed LDH₂₀₀₀ and LDH₃₅₀ by stirring in the aqueous NaHCO₃ solutions at 0.2 eq and 3 eq for 0.5 h and 12 h.

Fig. S3 and S4 indicated the AFM images and height profile of LDH reconstructed with MO at representative concentration and time point. As we could clearly observe in the profile, the surface of reconstructed LDH was rougher than pristine; the surface roughness becomes more significant with increasing MO concentration in eq unit (which is also strongly related to loading ratio) as well as reaction time.

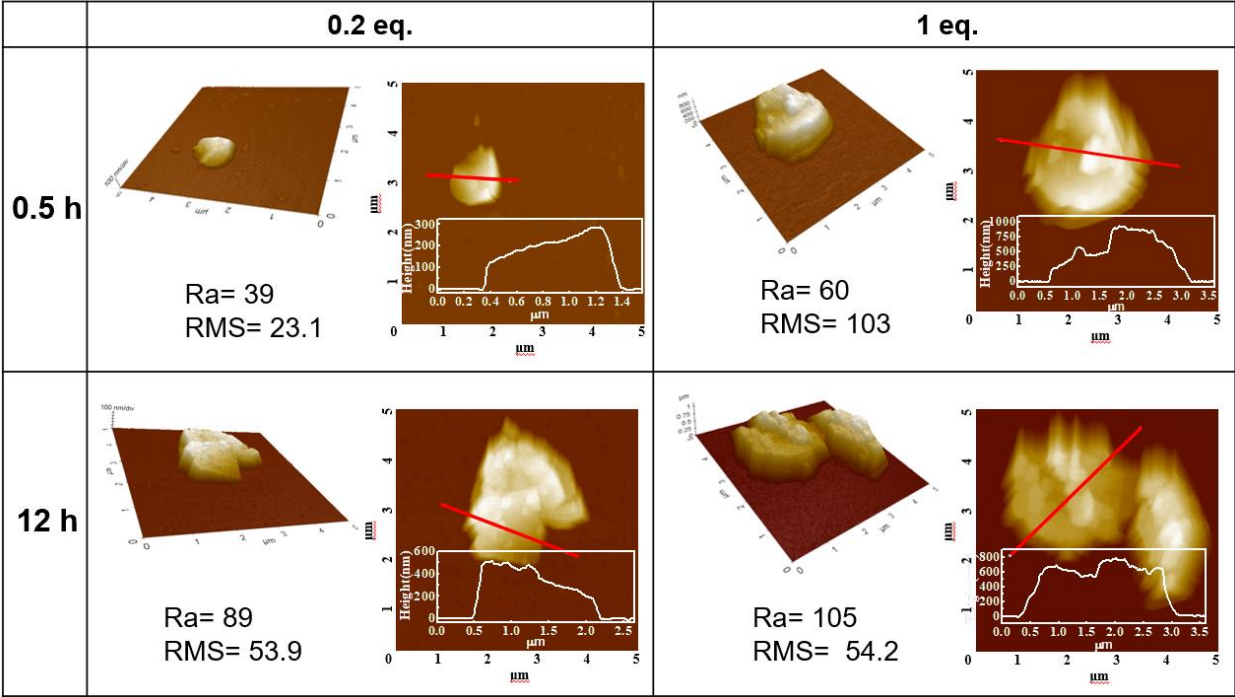


Fig. S3 Atomic force microscopic (AFM) images of the reconstructed LDH₂₀₀₀ by stirring in the aqueous MO solutions at 0.2 and 1 eq for 0.5 and 12 h.

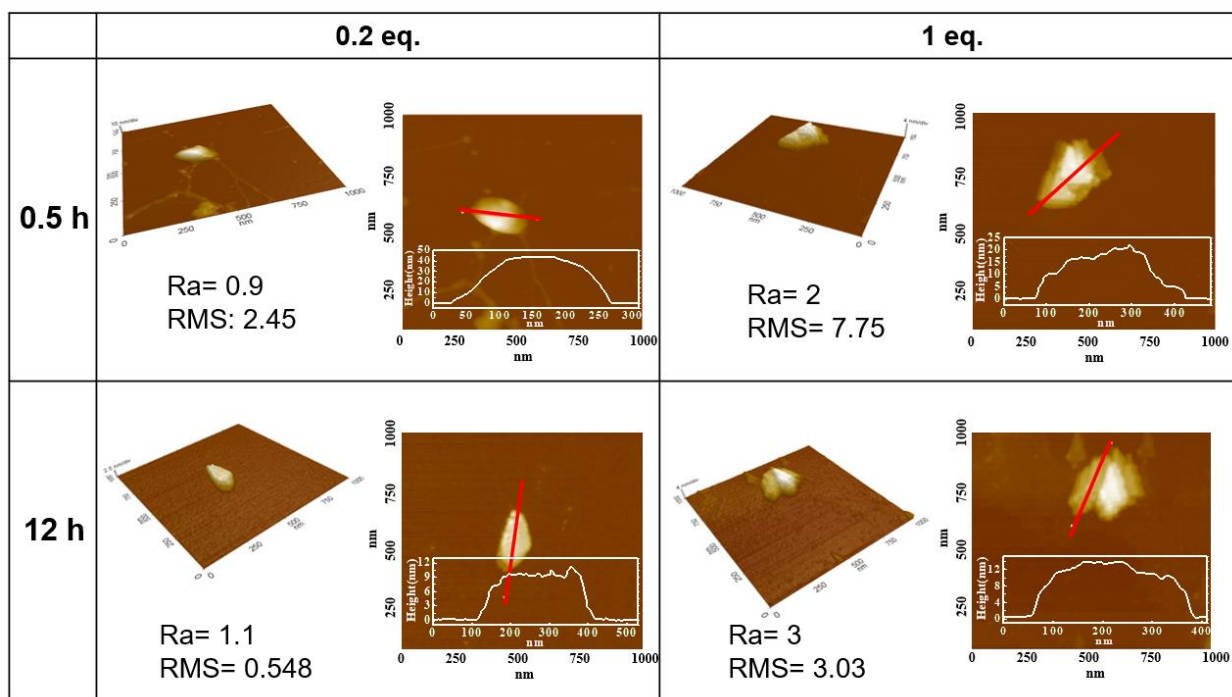


Fig. S4 Atomic force microscopic (AFM) images of the reconstructed LDH₃₅₀ by stirring in the aqueous MO solutions at 0.2 and 1 eq for 0.5 and 12 h.