Electronic Supplementary Information

Efficient, scalable, closed-loop synthesis of highly crystalline pure phase MgAl-layered double hydroxides intercalated with hydroxyl anions

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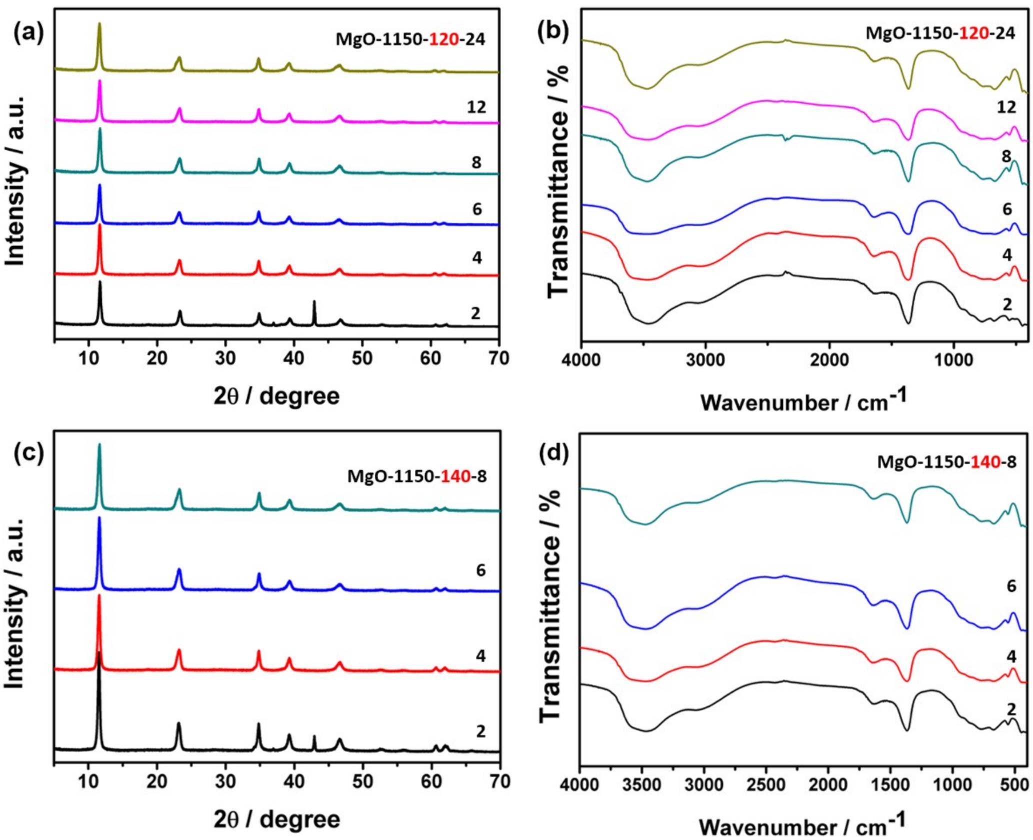

Fig. S1 XRD patterns and FTIR spectra of MgO-1150-T-t.

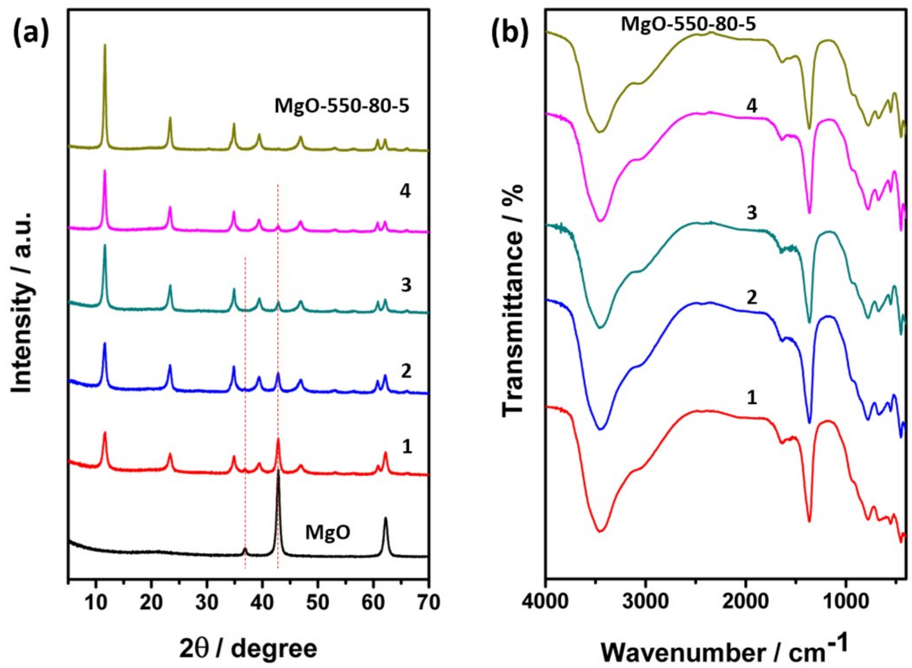

Fig. S2 XRD patterns and FTIR spectra of MgO-550-80-t.

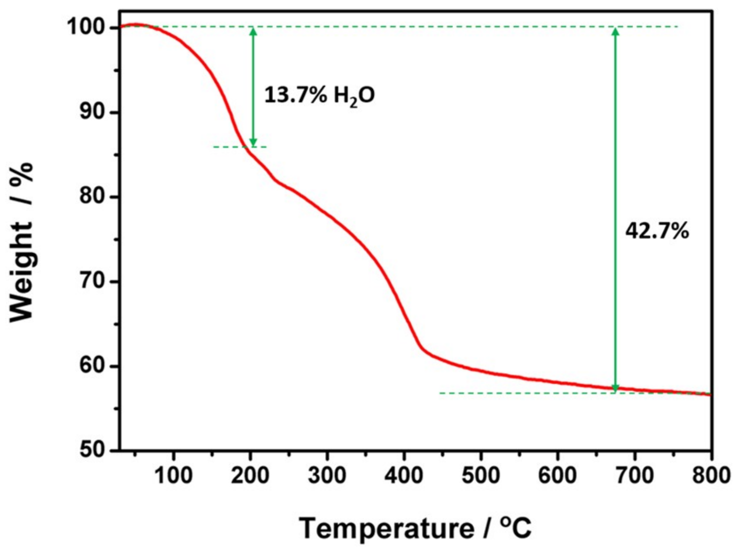

Fig. S3 TGA of MgO-550-80-5. The first weight loss step (30-180 ^oC) is assigned to the removal of water molecules in the interlayer of MgAl-LDH. The contents of Mg and Al in LDH were detected by ICP, and the Mg/ Al ratio is 2.57:1. The carbon content determined via CHN is 0.3 wt%. Consequently, the chemical formula of MgO-550-80-5 was estimated to be $[Mg_{0.720}Al_{0.280}(OH)_2](OH)_{0.242}(CO_3)_{0.019}\bullet 0.57H_2O$. The total weight loss (42.7%) is consistent with the calculated value (42.0%) on the basis of the empirical formula.

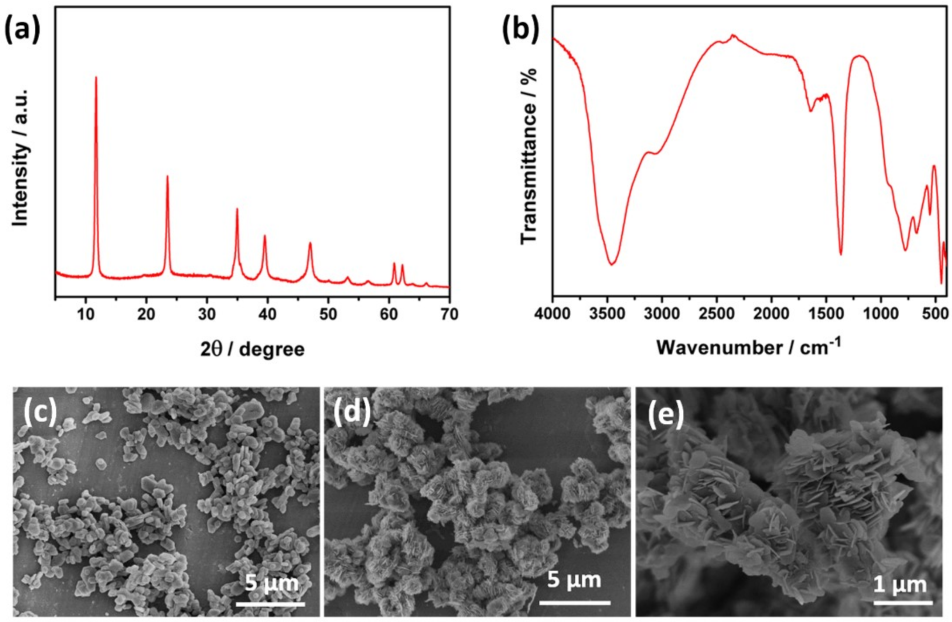

Fig. S4 MgAl-LDH from regenerative $NaAlO_2$. (a) XRD pattern and (**b**) FTIR spectrum of MgO-550-80-5, (**c**) SEM image of pristine MgO-550, (**d** and **e**) SEM images of MgO-550-80-5.

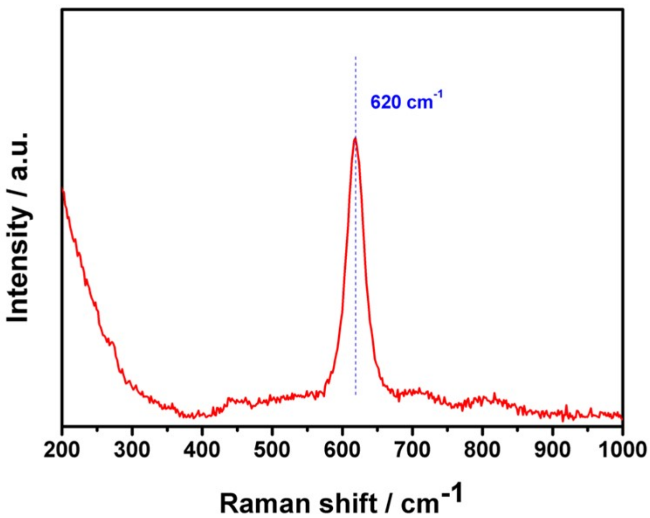

Fig. S5 Raman spectrum of $NaAlO_2$ solution (1.5 M). The peak at 620 cm^{-1} is assigned to symmetric stretching vibration[1] of tetrahedral $Al(OH)_4^-$.

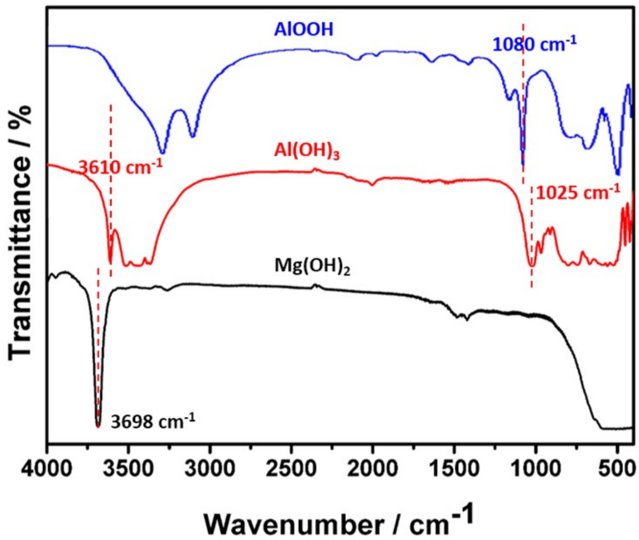

Fig. S6 FTIR spectra of $Mg(OH)_2$, $Al(OH)_3$, and AlOOH.

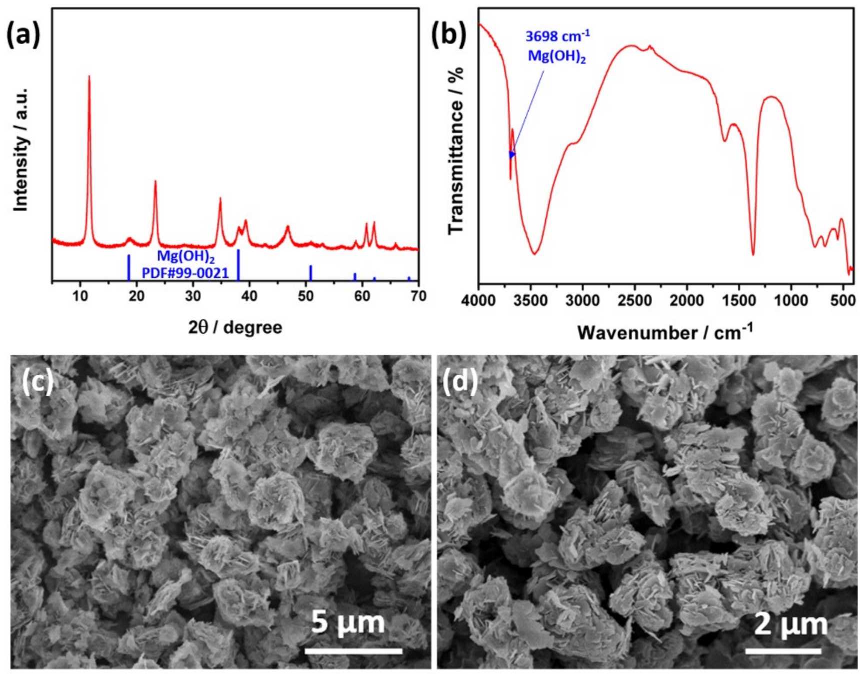

Fig. S7 (a) XRD pattern, (b) FTIR spectrum, and (c and d) SEM images of MgO-550-120-1. The trace of $Mg(OH)_2$ can be observed in XRD pattern and FTIR spectrum. The sharp absorption peak at 3698 cm^{-1} in FTIR spectrum is assigned to the stretching vibration of OH in $Mg(OH)_2$.

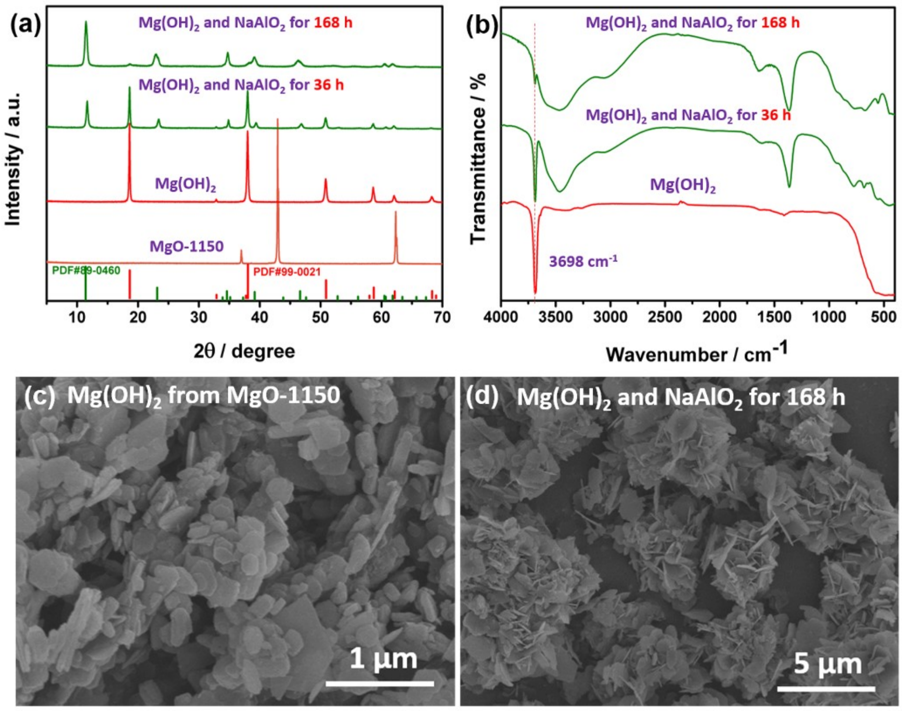

Fig. S8 Two-steps synthesis of MgAl-LDH from MgO [$Mg(OH)_2$] and $NaAlO_2$ at 100 ^oC. (a) XRD patterns, (b) FTIR spectra, (c) SEM image of $Mg(OH)_2$ from MgO-1150 at 100 ^oC, (d) SEM image of the precipitates after 168 h. MgO-1150 was converted completely into $Mg(OH)_2$ by hydrothermal treatment at 100 ^oC for 12 h. Then, $NaAlO_2$ was added to the hydrothermal system. According to the Le chatelier's law, the dissolution-precipitation equilibrium of $Mg(OH)_2$ shifts because of the removal of Mg^{2+} for constructing MgAl-LDH in the presence of $Al(OH)_4^-$. Indeed, the MgAl-LDH phase presented in the precipitates. However, a great deal of $Mg(OH)_2$ survived even after 36 h. Although MgAl-LDH is more thermodynamically stable than $Mg(OH)_2$,[2] the dissolution rate of $Mg(OH)_2$ was suppressed dramatically in an alkaline condition (pH > 11.7, 25℃) owing to the "common-ion effect". After 168 h, a trace of $Mg(OH)_2$ still can be observed, which is much slower than that of MgO. Given the sluggish kinetics of $Mg(OH)_2$ compared with MgO, it can be inferred that the $Mg(OH)_2$ is not the essential intermediate during DIDR, otherwise $Mg(OH)_2$ would be detected in FTIR spectra and XRD patterns.

Table S1 Specific surface areas of commercial products of MgAl-LDH and MgO-550-80-5.

	Company	Type	Specific area/ $m^2\ g^{-1}$
1	Sakai Chemical Industry Co., Ltd.	HT-1	**6-12**
2	Kyowa Kagaku Kogyo Co., Ltd.	DHT-4	**10-15**
3	Hunan Shaoyang Tiantang Auxiliaries Chemical Co., Ltd.	-	**8-20**
4	HEOWNS	-	**20**
5	This work	MgO-550-80-5	**56.3**

References

1. Johnston, C. T.; Agnew, S. F.; Schoonover, J. R.; Kenney, J. W.; Page, B.; Osborn, J.; Corbin, R, *Environ. Sci. Technol.* 2002, **36**, 2451-2458.
2. Johnson, C. A.; Glasser, F. P, *Clays Clay Miner.* 2003, **51**, 1-8.