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Supporting Information

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Experimental and DFT study of F^- removed by Cl^--hydrotalcite

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X-ray powder diffraction (XRD) pattern of Cl^--hydrotalcite was collected using an X-ray diffractometer (Bruker D8 Advance, Germany) with a copper K α source (λ =0.15418 nm) at a constant scanning rate (5° min^{-1}) over the 2θ range 10°-90°. Test voltage was 40 kV and test current was 40 mA.

The XRD spectrum of Cl-LDH is shown in Fig. S1. The diffraction peak of the (0 0 3) crystal face of hydrotalcite is the strongest, the diffraction peaks of (0 0 6) and (0 0 9) crystal faces of hydrotalcite are also strong. These diffraction peaks all belong to characteristic peaks of hydrotalcite.

MSD was calculated by Equation (1).

$$\mathrm{MSD(t)} = \left[\frac{1}{\mathrm{N}}\sum_{\mathrm{i}=1}^{\mathrm{N}}|\mathrm{R_i(t)} - \mathrm{R_i(0)}|^2\right] \quad (1)$$

The diffusion coefficient (D) of F^- was estimated by Einstein equation, as shown in Equation (2):

$$\mathrm{D} = \frac{1}{2\mathrm{dN}}\lim_{\mathrm{t}\to\infty}\frac{\mathrm{d}}{\mathrm{dt}}\sum_{\mathrm{i}=1}^{\mathrm{N}}\langle[\mathrm{R_i(t)} - \mathrm{R_i(0)}]^2\rangle \quad (2)$$

Here, d is the dimension of the system and N is the number of target molecules in the system. Ri(t) and Ri(0) are the coordinates of fluorine at time T and time 0, respectively. Plot the obtained MSD data with time and fit the straight line. The slope a is $\frac{\mathrm{d}}{\mathrm{dt}}\sum_{\mathrm{i}=1}^{\mathrm{N}}\langle[\mathrm{R_i(t)} - \mathrm{R_i(0)}]^2\rangle$. Since MSD has averaged the ion number N, the diffusion coefficient can be simplified as:

$$\mathrm{D} = \frac{\mathrm{a}}{6} \quad (3)$$

In this article, the Fukui functions are defined by the following formula:

$$\mathrm{f(r)} = \left[\frac{\partial\rho(\mathrm{r})}{\partial\mathrm{N}}\right]_{\mathrm{V}} \quad (4)$$

In the above equation, ρ(r) is the electron density function, N is the total number of electrons in the system, and V is the external potential energy. In this study, the electrophilic Fukui function (f^-) and the nucleophilic Fukui function (f^+) are used, and their expressions are as follows:

$$\mathrm{f^-(r)} = \rho_{\mathrm{N}}(\mathrm{r}) - \rho_{\mathrm{N-1}}(\mathrm{r}) \quad (5)$$

$$\mathrm{f^+(r)} = \rho_{\mathrm{N+1}}(\mathrm{r}) - \rho_{\mathrm{N}}(\mathrm{r}) \quad (6)$$

Here, $\rho_N(r)$ is the density function under normal conditions. $\rho_{N-1}(r)$ and $\rho_{N+1}(r)$ are the density functions of the system with one unit of positive charge and one unit of negative charge, respectively. The greater the isosurface of the value of f^- and f^+ near the atom is, the easier it is for the electrophilic reaction and nucleophilic reaction to occur on the atom.

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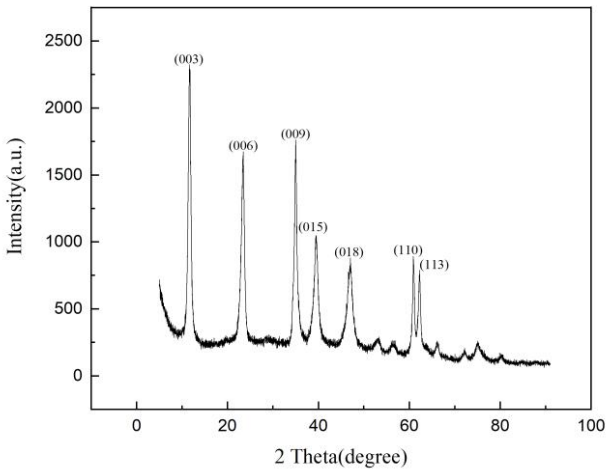

Fig. S1 XRD pattern of Cl^--hydrotalcite.

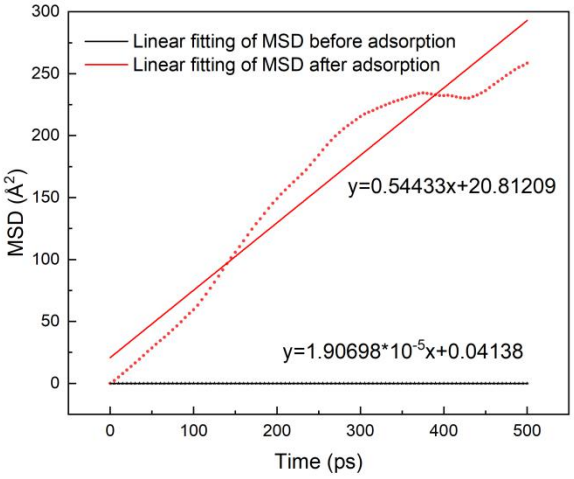

Fig. S3 MSD plot of F^- in water and in hydrotalcite solution.

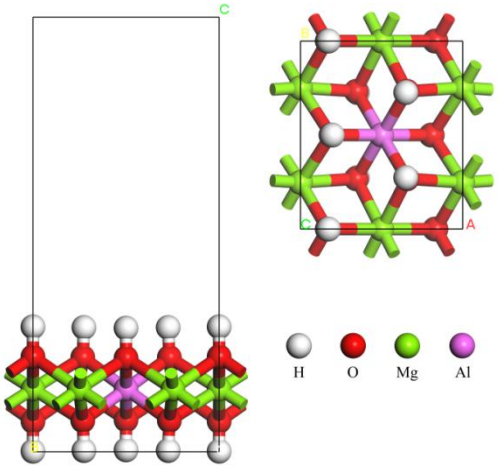

Fig. S2 Model of hydrotalcite.

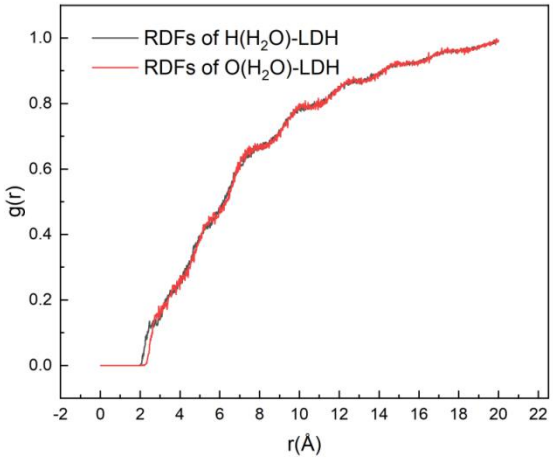

Fig. S4 RDFs of $H(H_2O)$-LDH and $O(H_2O)$-LDH.

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Table S1 The Fukui functions and dual descriptors of the hydrotalcite calculated at DFT/GGA/DNP

	f^+_{LDH}	f^-_{LDH}	f^+_{F-LDH}	f^-_{F-LDH}	f^+_{Cl-LDH}	f^-_{Cl-LDH}	Δf_{LDH}	Δf_{F-LDH}	Δf_{Cl-LDH}
O(1)	-0.056	0.015	-0.009	0.049	-0.016	0.004	-0.071	-0.058	-0.020
H(2)	0.187	0.072	0.045	0.054	0.053	0.042	0.115	-0.009	0.011
O(3)	-0.056	0.015	-0.100	0.006	-0.094	-0.002	-0.071	-0.106	-0.092
H(4)	0.187	0.072	0.334	0.068	0.320	0.028	0.115	0.266	0.292
O(5)	-0.056	0.015	-0.100	0.006	-0.094	-0.002	-0.071	-0.106	-0.092
H(6)	0.187	0.072	0.335	0.068	0.320	0.028	0.115	0.267	0.292
O(7)	-0.056	0.015	-0.009	0.049	-0.016	0.005	-0.071	-0.058	-0.021
H(8)	0.187	0.072	0.045	0.054	0.053	0.042	0.115	-0.009	0.011
Mg(9)	0.008	0.040	-0.002	0.043	0.001	0.017	-0.032	-0.045	-0.016
Mg(10)	0.008	0.040	-0.002	0.043	0.001	0.017	-0.032	-0.045	-0.016
Mg(11)	0.007	0.039	-0.005	0.041	-0.002	0.017	-0.032	-0.046	-0.019
O(12)	-0.053	0.074	-0.025	0.036	-0.028	0.000	-0.127	-0.061	-0.028
H(13)	0.171	0.087	0.091	0.078	0.097	0.036	0.084	0.013	0.061
O(14)	-0.053	0.074	-0.091	0.032	-0.087	0.006	-0.127	-0.123	-0.093
H(15)	0.171	0.087	0.287	0.076	0.277	0.030	0.084	0.211	0.247
Al(16)	-0.040	0.026	-0.063	-0.003	-0.062	-0.020	-0.066	-0.06	-0.042
O(17)	-0.056	0.022	-0.010	0.049	-0.015	0.006	-0.078	-0.059	-0.021
H(18)	0.184	0.073	0.041	0.055	0.043	0.032	0.111	-0.014	0.011
O(19)	-0.056	0.022	-0.095	0.014	-0.090	-0.001	-0.078	-0.109	-0.089
H(20)	0.188	0.073	0.324	0.072	0.313	0.029	0.115	0.252	0.284
F^-			0.008	0.111				-0.103	
Cl^-					0.025	0.686			-0.661

(Note: The f^+ and f^- of F^- and Cl^- are both 1 before adsorption, respectively.)

Table S2 Mulliken charges of atoms before and after F^- or Cl^- adsorption on the surface of hydrotalcite

System	F^-	Cl^-	LDH	Cl-LDH	F-LDH
-OH(1,2)			-0.466	-0.489	-0.510
-OH(3,4)			-0.466	-0.476	-0.481
-OH(5,6)			-0.466	-0.476	-0.481
-OH(7,8)			-0.466	-0.489	-0.510
Mg(9)			1.088	1.078	1.079
Mg(10)			1.088	1.078	1.079
Mg(11)			1.093	1.083	1.084
-OH(12,13)			-0.512	-0.523	-0.526
-OH(14,15)			-0.512	-0.521	-0.524
Al(16)			1.546	1.531	1.518
-OH(17,18)			-0.464	-0.483	-0.503
-OH(19,20)			-0.464	-0.474	-0.478
F^-	-1				-0.745
Cl^-		-1		-0.841	

Table S3 Hirshfeld charges of atoms before and after F^- or Cl^- adsorption on the surface of hydrotalcite

System	F^-	Cl^-	LDH	Cl-LDH	F-LDH
O(1)			-0.2483	-0.2665	-0.2798
H(2)			0.1905	0.0994	0.0897
O(3)			-0.2483	-0.2523	-0.2546
H(4)			0.1905	0.1809	0.1747
O(5)			-0.2483	-0.2523	-0.2546
H(6)			0.1905	0.1809	0.1747
O(7)			-0.2483	-0.2665	-0.2799
H(8)			0.1905	0.0994	0.0897
Mg(9)			0.3826	0.3739	0.3701
Mg(10)			0.3826	0.3739	0.3701
Mg(11)			0.3910	0.3820	0.3777
O(12)			-0.2931	-0.2950	-0.2945
H(13)			0.1752	0.1517	0.1569
O(14)			-0.2931	-0.2957	-0.2971
H(15)			0.1752	0.1659	0.1600
Al(16)			0.4294	0.4127	0.4096
O(17)			-0.2493	-0.2675	-0.2777
H(18)			0.1904	0.0971	0.0927
O(19)			-0.2493	-0.2524	-0.2540
H(20)			0.1904	0.1814	0.1762
F^-	-1				-0.4493
Cl^-		-1		-0.5506	