Supplemental info

Table 1. Supporting information contains: RI-SCS-MP2/aug-cc-pVTZ(COSMO)//B3LYP/6-311++G(d,p) calculated total energies and solvation energiers, as well as calculated gas and solvated (COSMO) dipole moments, μ (in Debye, D) and isotropic polarizability, <α> (in Hartrees^3), from the reaction of H_2O and CO_2 to form carbonic acid[a]

Clusters and molecules	Total energy, Hartrees	Solvation energy, kcal/mol[b]	μ(D), gas phase	μ(D), COSMO	<α>(Hartrees^3), gas phase	<α>(Hartrees^3), COSMO
Reactants[c]						
H_2O	-76.315929	-6.95	1.977	2.409	6.526	7.189
H_2O dimer	-152.632018	-11.66	3.042	3.395	17.088	20.406
H_2O trimer	-228.945827	-10.83	1.015	1.274	25.550	30.833
CO_2	-188.293374	-1.88	0.000	0.000	15.774	19.349
$Al(OH)_3$	-469.365483	-32.24	0.010	0.010	30.185	37.197
Intermediates[d]						
CO_2+H_2O	-264.608304	-6.80	2.386	2.649	24.041	29.406
CO_2+H_2O TS	-264.524934	-7.29	2.826	3.601	25.746	32.732
CO_2+2H_2O	-340.923473	-8.96	1.863	2.249	32.706	39.752
CO_2+2H_2O TS	-340.875576	-14.25	5.454	6.759	34.242	43.149
CO_2+3H_2O	-417.240397	-10.95	1.434	1.704	41.465	50.217
CO_2+3H_2O TS	-417.204046	-19.27	7.103	8.597	43.256	53.907
Products[d]						
H_2CO_3	-264.592946	-10.37	3.378	4.252	23.561	29.285
H_2CO_3+H_2O	-340.912089	-13.24	5.460	6.585	32.190	39.650
H_2CO_3+2H_2O	-417.229944	-15.33	4.904	5.911	41.113	50.424

a. Calculated using COSMO solvation with water as a solvent and ZPE gas phase corrected. b. calculated by subtracting total RI-SCS-MP2/aug-cc-pVTZ energies of the gas phase calculations from those of using COSMO solvation model. c,d. Clusters and molecules shown in Figures 1 and 2, respectively.

Table 2. Supporting information contains: RI-SCS-MP2/aug-cc-pVTZ(COSMO)//B3LYP/6-311++G(d,p) calculated total energies and solvation energiers, as well as calculated gas and solvated (COSMO) dipole moments, μ (in Debye, D) and isotropic polarizability, <α> (in Hartrees^3), from the reaction of H_2O and CO_2 to form carbonic acid in the presence of $Al(OH)_3$[a]

Reaction Coordinate	Total energy, Hartrees	Solvation energy, kcal/mol[b]	μ(D), gas phase	μ(D), COSMO	<α>(Hartrees^3), gas phase	<α>(Hartrees^3), COSMO
CO_2+H_2O[c]						
I	-733.978261	-19.13	2.112	2.359	53.175	65.742
II	-733.958526	-18.08	1.086	1.333	53.975	66.447
III	-733.971878	-23.17	4.333	5.277	52.862	65.446
IV (TS)	-733.921983	-19.70	4.339	5.352	52.893	65.922
V	-733.979512	-20.22	2.197	2.719	52.357	64.991
VI	-733.961032	-28.02	6.450	7.162	53.093	65.327
CO_2+2H_2O[c]						
I	-810.301015	-17.22	2.225	2.651	61.573	N/A[d]
II	-810.276104	-19.82	1.595	1.812	62.507	76.620
III	-810.291151	-23.19	3.630	4.419	61.449	75.707
IV (TS)	-810.276762	-22.47	3.849	4.723	62.172	77.139
V	-810.297527	-21.50	2.942	3.598	61.049	75.337
VI	-810.284677	-30.80	9.752	10.581	61.930	75.687
CO_2+3H_2O[c]						
I	-886.619898	-16.51	1.303	1.501	69.319	N/A[d]
II	-886.587763	-21.22	4.191	4.833	71.162	87.133
III	-886.613092	-25.45	3.815	4.763	70.611	86.498
IV (TS)	-886.601285	-26.33	5.710	6.709	71.664	88.609
V	-886.618006	-23.37	3.261	3.832	70.048	86.083
VI	-886.601686	-32.62	9.661	10.459	70.816	86.370

a. Calculated using COSMO solvation with water as a solvent and ZPE gas phase corrected. b. calculated by subtracting total RI-SCS-MP2/aug-cc-pVTZ energies of the gas phase calculations from those of using COSMO solvation model. c. Cluster models shown in Figure 4. d. polarizability calculations failed to converge