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Supplementary Materials for

A scalable solid-state nanoporous network with atomic-level interaction design for carbon dioxide capture

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This PDF file includes:

Fig. S1. Scalable MNNs^{Cya}⊃DETA samples (1 kg) after grinding via mortar and pestle.

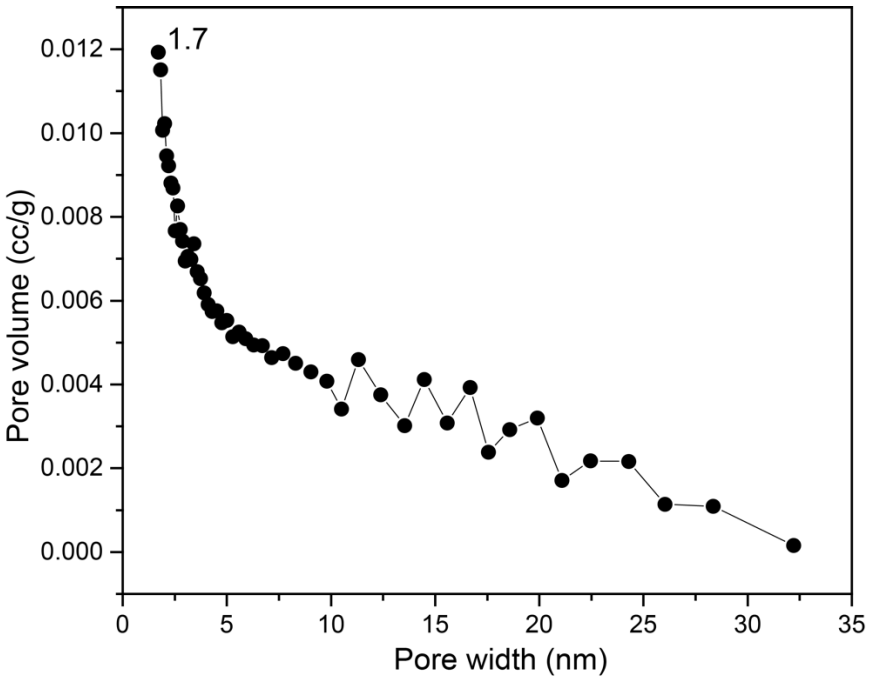

Fig. S2. Pore size distribution of MNNs^{Cya}⊃DETA.

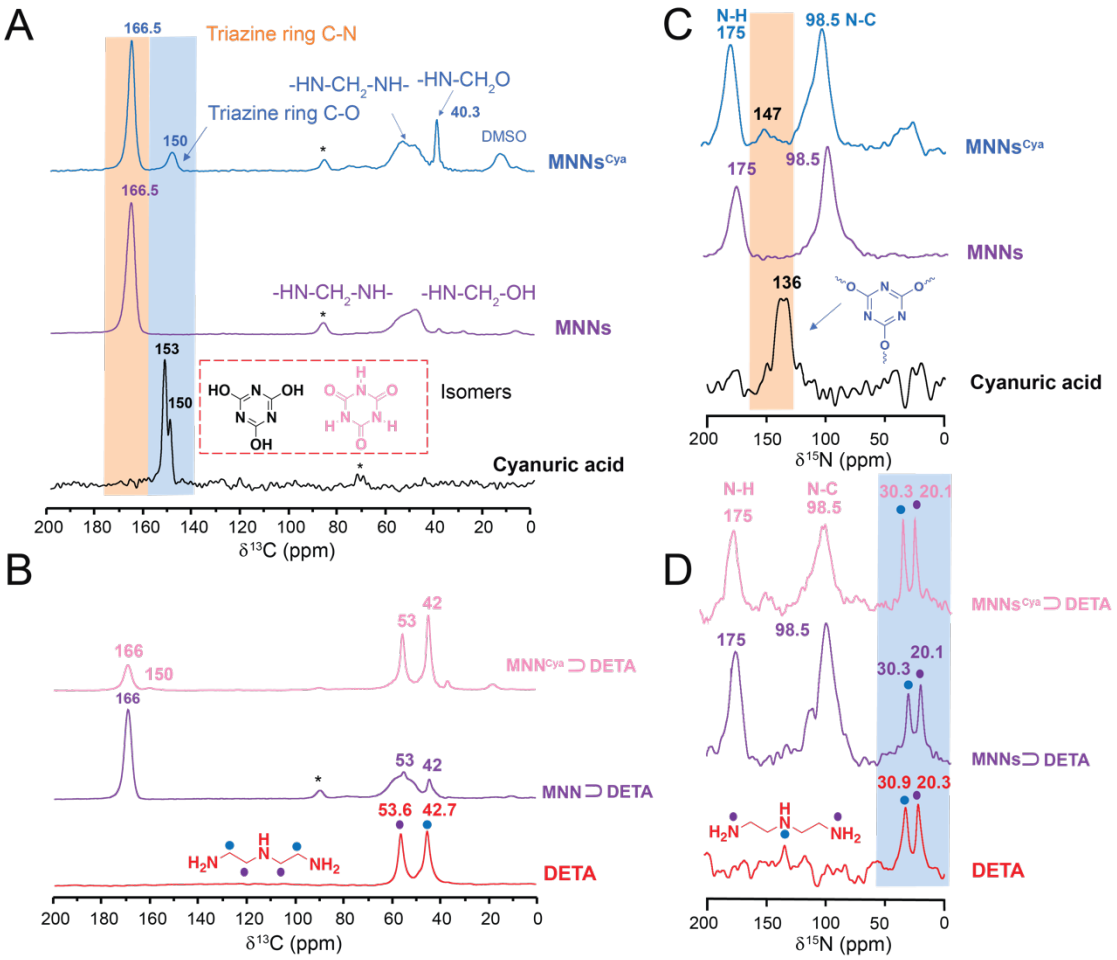

Fig. S3. Determination of chemical structures of DETA and cyanuric acid-modified MNNs by ^{13}C and ^{15}N MAS NMR spectra. A, Direct excitation ^{13}C MAS NMR spectra of cyanuric acid, MNNs, and MNNs^{Cya}. **B**, ^{13}C CP-MAS NMR spectra of DETA, MNNs ⊃ DETA, and MNNs^{Cya} ⊃ DETA. **C**, ^{15}N CP-MAS NMR spectra of cyanuric acid, MNNs, and MNNs^{Cya}. **D**, ^{15}N CP-MAS NMR spectra of DETA, MNNs ⊃ DETA, MNNs^{Cya} ⊃ DETA.

Deconvolution

To identify the structure of MNNs^{Cya} and MNNs^{Cya} ⊃ DETA, quantitative ^{13}C NMR spectral fitting was carried out using the DMfit software. Deconvolutions were calculated using a mixture of Gaussian and Lorentzian lineshapes to describe the different features in the spectra. Fits were repeated up to four times for each series of data in order to estimate errors. Example deconvolutions of ^{13}C NMR spectra are depicted in **Fig. S4**.

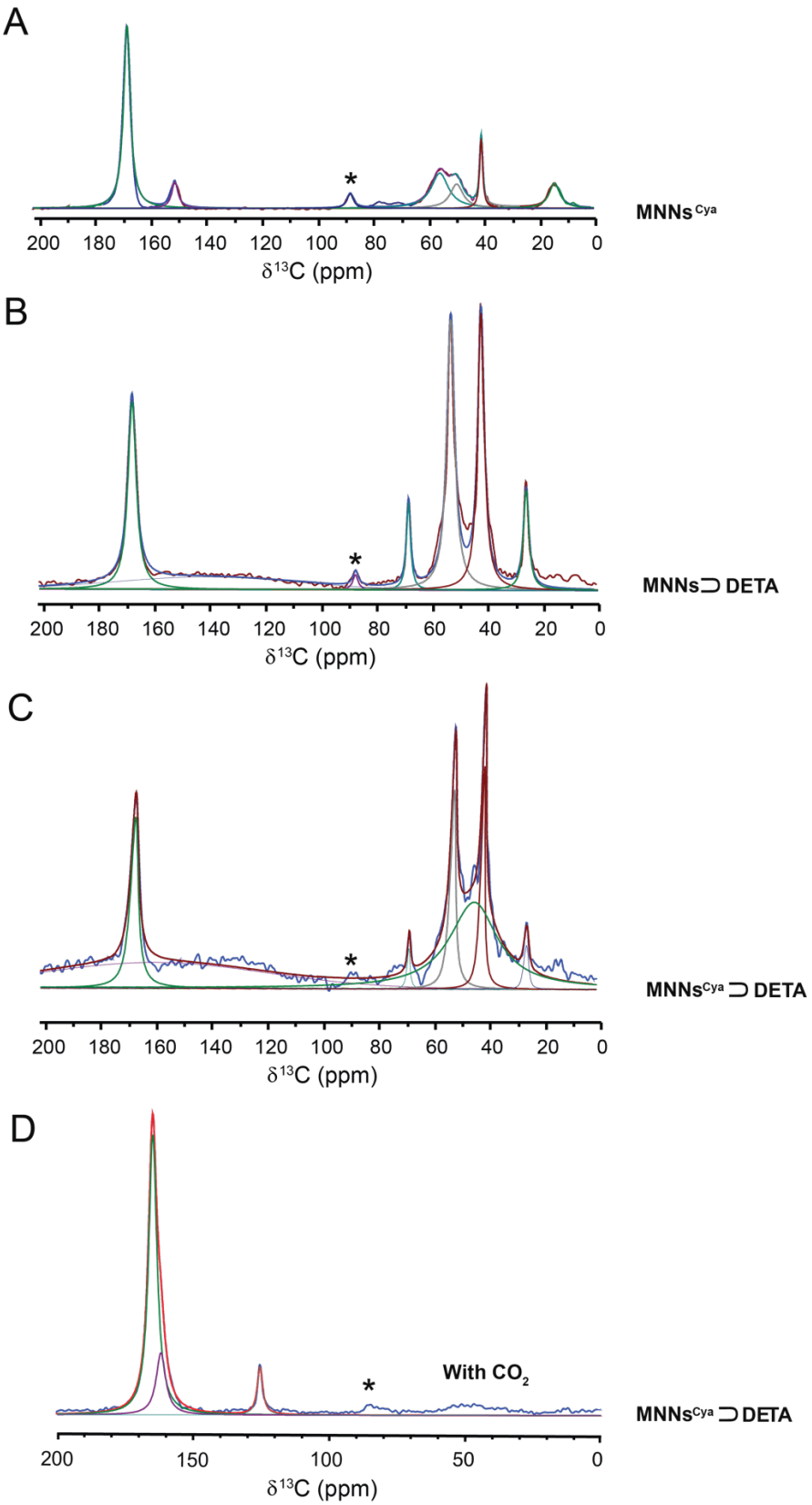

Fig. S4. Representative deconvolutions of ^{13}C NMR spectra recorded for MNNs^{Cya} ⊃ DETA, MNNs^{Cya}, and MNNs ⊃ DETA. Experimental lineshapes are shown in blue, while the sum of individually fitted spectra is shown in red. Asterisks denote spinning side bands.

Table S1. Spectral simulation parameters of Gaussian/Lorentzian of $MNNs^{Cya}$, $MNNs^{Cya}$ ⊃ DETA, MNNs ⊃ DETA, and $MNNs^{Cya}$ ⊃ DETA-CO_2 using dmfit software.

	^{13}C NMR spectra		
	δ (ppm)	**Gaussian/ Lorentzian**	**Proportion**
$MNNs^{Cya}$	166.5	0.95	44.3 %
	150.0	0.80	6.8 %
	55.0	0.32	15.9%
	49.0	0.28	12.1%
	40.3	0.90	13.0%
	14.1	0.3	7.9%
MNNs ⊃ DETA	166.5	0.95	21.3 %
	67.8	0.98	13.2 %
	53.0	0.92	26.4%
	42.0	0.95	24.1%
	25.7	0.90	15.0%
$MNNs^{Cya}$ ⊃ DETA	166.5	0.98	20.3 %
	151.0	0.15	4.0 %
	67.5	0.92	16.4%
	53.0	0.95	25.1%
	42.0	0.90	25.3%
	25.7	0.90	8.9%
$MNNs^{Cya}$ ⊃ DETA-CO_2	164.7	0.98	73.2%
	161.1	0.95	18.7%
	125.4	0.95	8.1%

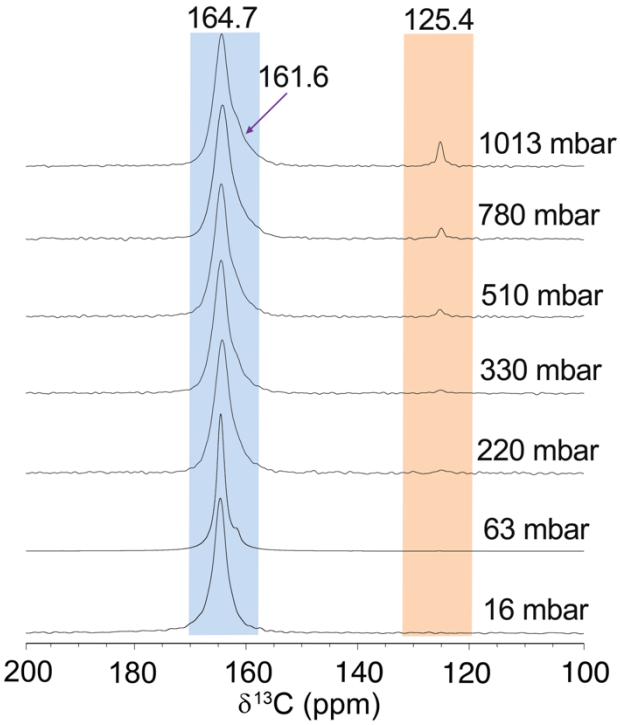

Fig. S5. ^{13}C MAS (15 kHz) NMR (16.4 T) spectrum of MNNs^{Cya}⊃DETA dosed with $^{13}CO_2$ at room temperature (298 K) acquired by direct excitation. Each sample was rapidly dosed with $^{13}CO_2$ using the designed setup and allowed to equilibrate for at least 24 hours before measurement.

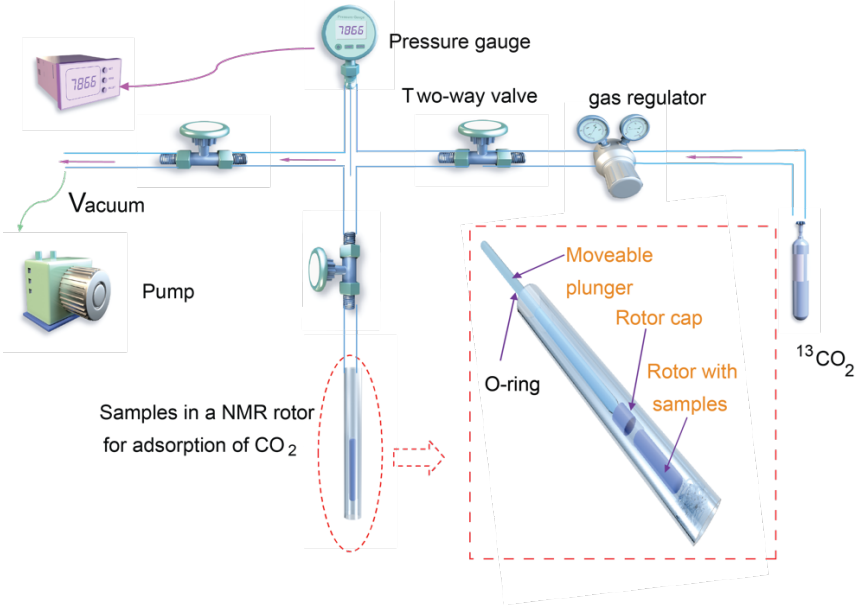

Fig. S6. Schematic of home-built setup for gas dosing of NMR samples. $^{13}CO_2$ gas was dosed at approximately 1 bar.

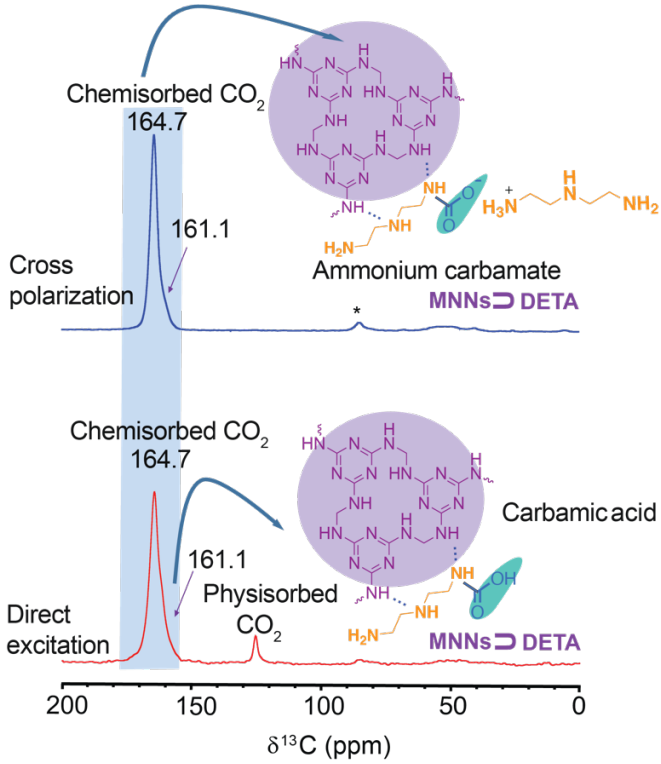

Fig. S7. Direct excitation ^{13}C MAS NMR spectra and cross-polarized ^{13}C MAS NMR spectra (with continuous-wave ^{1}H decoupling) of CO_2 adsorbed MNNs⊃ DETA.

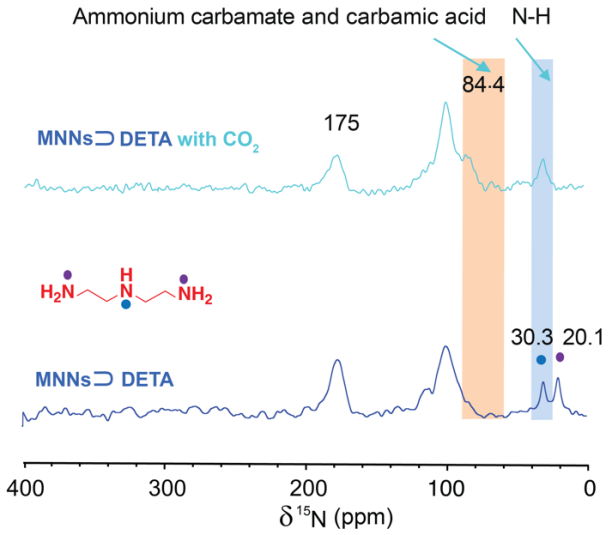

Fig. S8. Cross-polarized ^{15}N MAS NMR spectra of MNNs⊃ DETA with and without CO_2 adsorption.

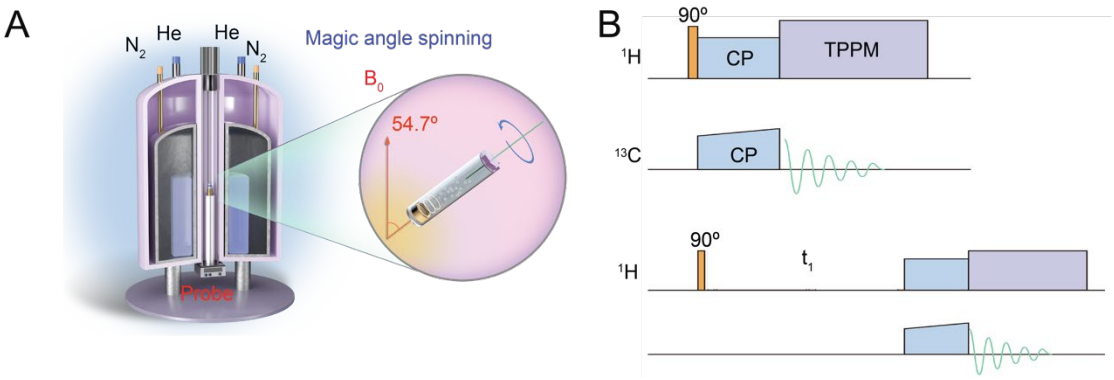

Fig. S9. Principle of solid-state nuclear magnetic resonance (NMR) spectroscopy. **(A)** Schematic diagram of solid-state NMR measurements. (**B**) Cross-polarization (CP) pulse timing diagram. 2D ^{1}H-^{13}C HETCOR with ^{1}H homonuclear decoupling pulse timing diagram. In these pulse sequences, the heteronuclear decoupling scheme can be two-pulse phase modulation (TPPM) in the CP experiment in the HETCOR experiment. t_1 and t_2 refer to time-domain increments for 2D experiments, and 90° pulse is shown as filled narrow rectangles.

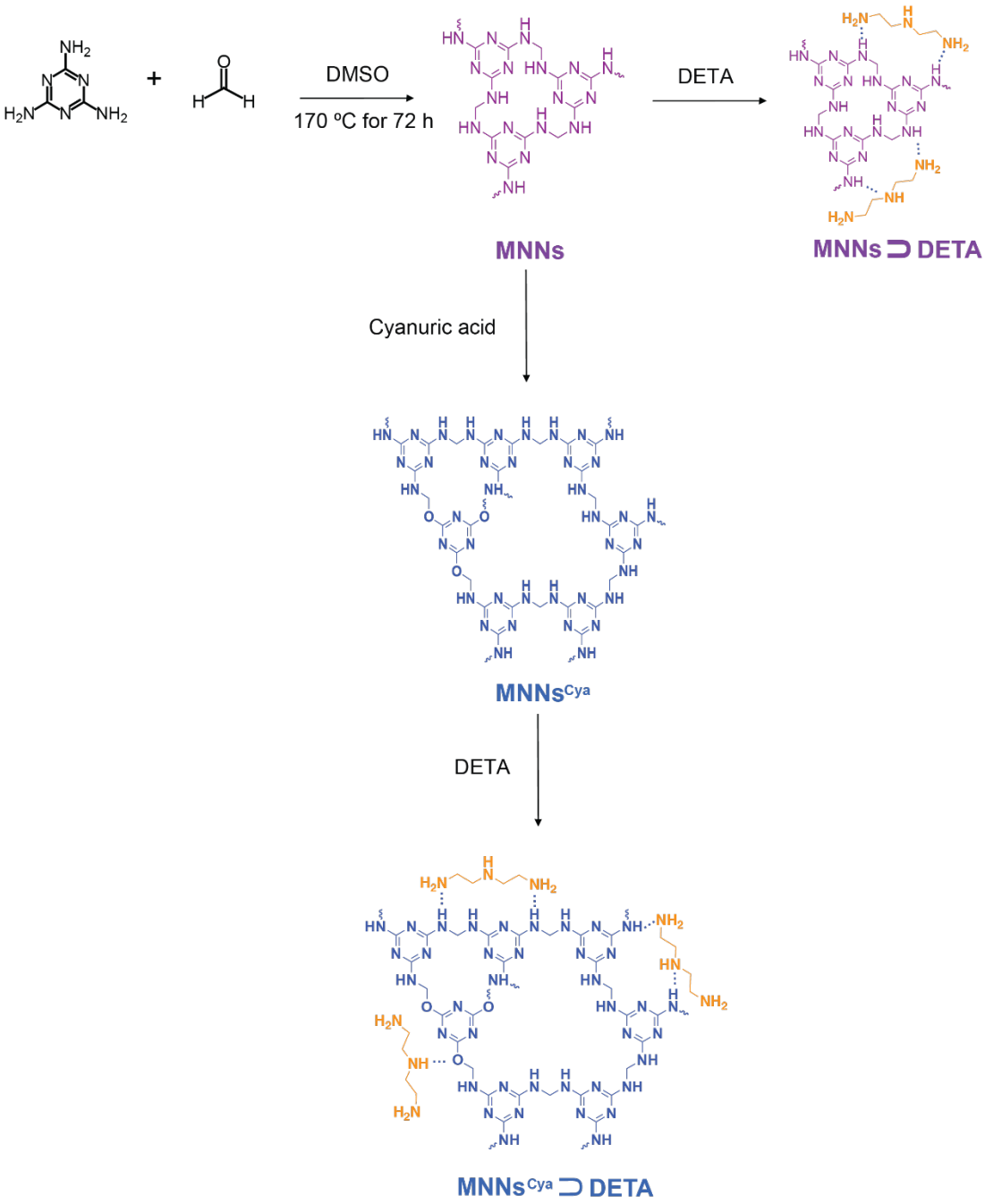

Fig. S10. The schematic synthesis of MNNs, MNNs^{Cya}, MNNs ⊃ DETA, and MNNs^{Cya} ⊃ DETA.

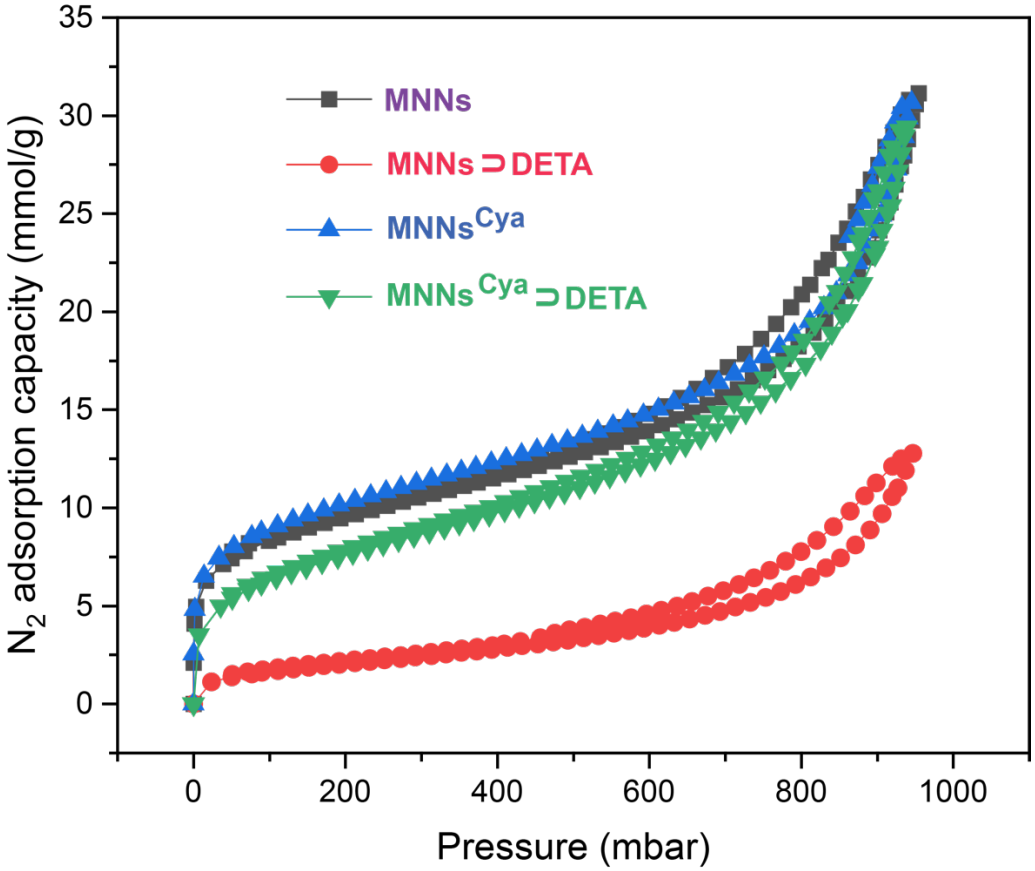

Fig. S11. N_2 adsorption and desorption isotherms for MNNs, MNNs ⊃ DETA, MNNs^{Cya}, and MNNs^{Cya} ⊃ DETA at 77 K.

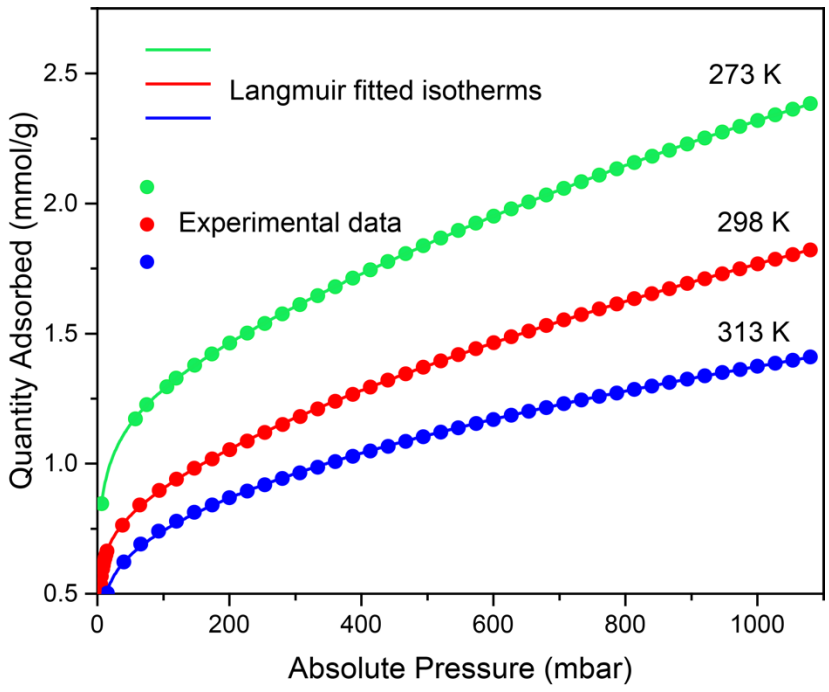

Fig. S12. Experimental data (symbols) and Langmuir modeling fit (solid curves) at 273 K (green), 298 K (red), and 313 K (blue) for CO_2 adsorption in MNNs^{Cya} ⊃ DETA.

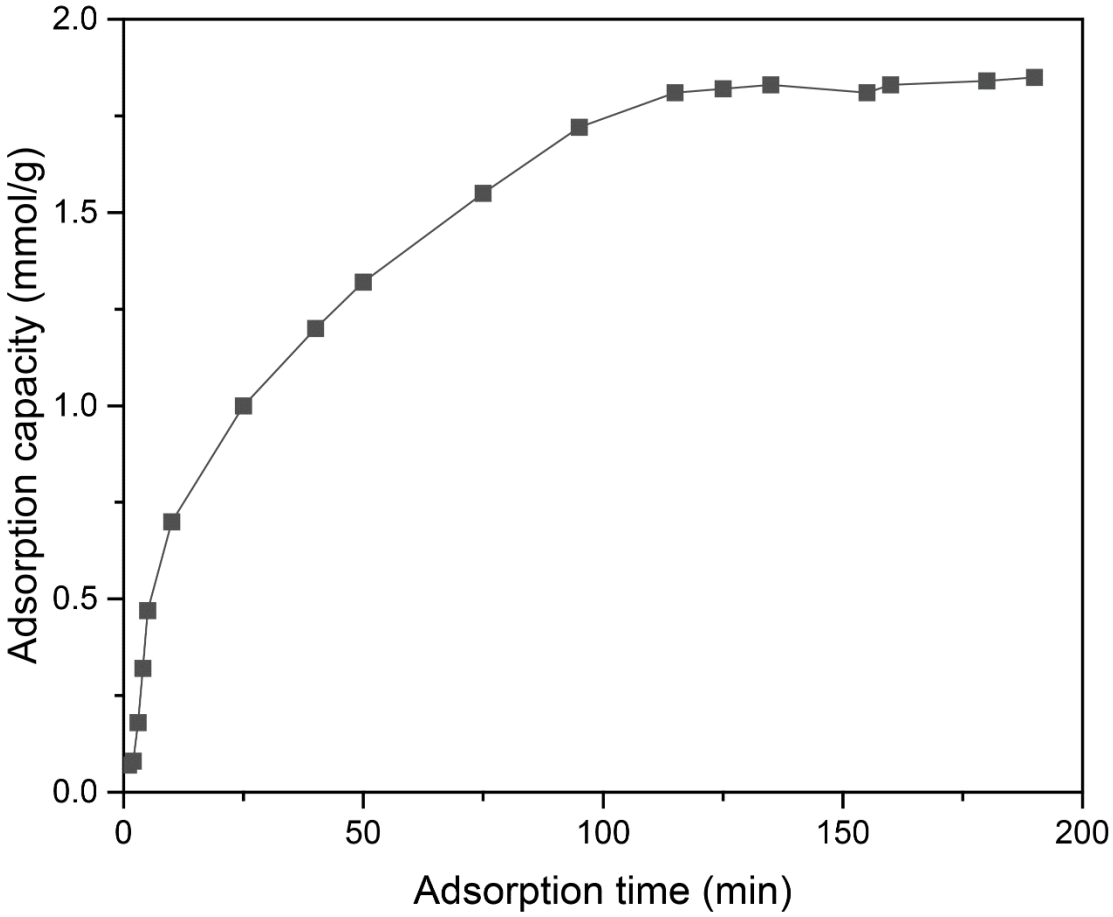

Fig. S13. Breakthrough adsorption experiments performed at 298 K with the feed containing CO_2 at the partial pressure of 1 bar and flow rate 0.1 L/min. The rate of CO_2 extraction is calculated as moles of CO_2 captured per gram adsorbent per unit time.

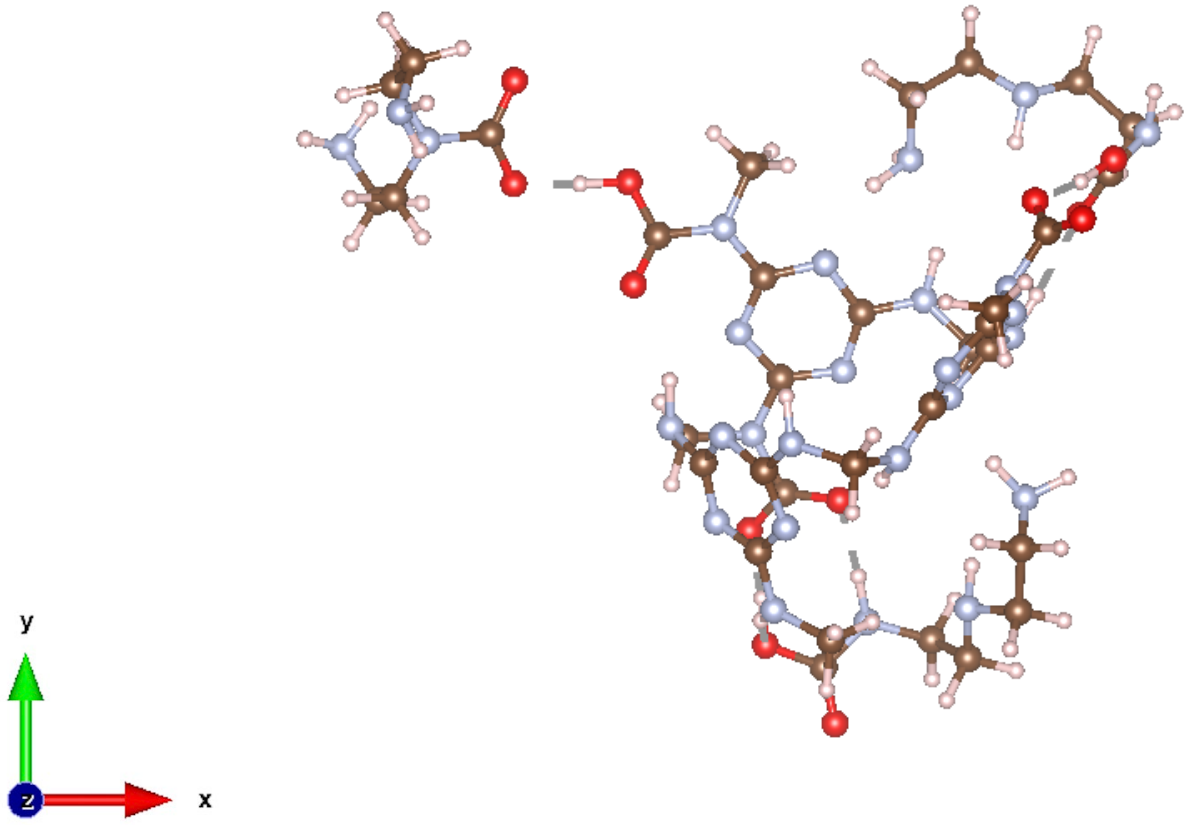

Fig. S14. Proposed DFT-calculated structure of CO_2-reacted mixed ammonium carbamate and carbamic acid within MNNs⊃DETA (Structure 1). The structure was obtained by DFT calculation at the TPSS-D3(BJ)/6-31G* level.

Coordinates:

	X	Y	Z
C	-1.303352	-4.577277	0.813264
N	-2.204985	-3.932952	0.042298
C	-2.486375	-2.676647	0.441392
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C	-1.174664	-2.840866	2.260456
N	-0.712272	-4.065482	1.927296
N	-0.588091	-2.252959	3.382043
N	-3.191880	-1.900041	-0.445678
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N	4.178870	0.562899	1.891947

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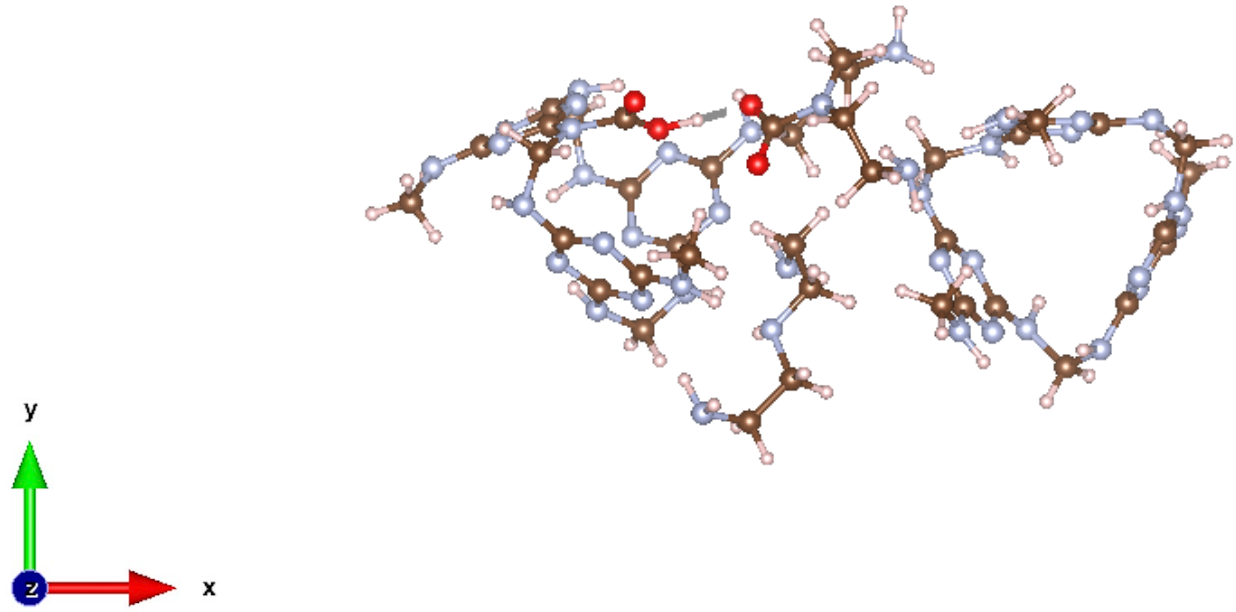

Fig. S15. DFT-calculated structure of CO_2-reacted mixed ammonium carbamate and carbamic acid within MNNs ⊃ DETA (Structure 2). The structure was obtained by DFT calculation at the TPSS-D3(BJ)/6-31G* level.

Coordinates:

	X	Y	Z
C	-8.742418	1.447104	-1.388897
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C	7.903970	-2.589298	-1.072235
N	8.215849	-1.908712	0.051890
C	8.880688	-0.758039	-0.184182
N	9.291008	-0.299949	-1.392196
N	9.129858	0.037226	0.907571
N	7.212778	-3.767148	-0.894749
N	9.167479	-0.626442	-3.687132
C	9.410990	1.475691	0.728892
N	8.552279	2.141652	-0.244355
C	7.178433	2.036465	-0.058828
N	6.462903	1.844491	-1.183343
C	5.123659	1.794205	-0.958493
N	4.501747	1.980626	0.220492
C	5.354792	2.082548	1.282865
N	6.713494	2.101156	1.203775
N	4.778303	2.157779	2.502421
N	4.368446	1.495791	-2.060973
C	2.950963	1.145490	-1.987781
N	2.579405	0.221752	-0.915111
C	3.217553	-0.967839	-0.702512
N	3.923347	-1.505610	-1.719842
C	4.517946	-2.677602	-1.396032
N	4.435825	-3.330746	-0.218871
C	3.692605	-2.675570	0.710857
N	3.076994	-1.490788	0.546350
N	5.308295	-3.248697	-2.369557
C	6.324359	-4.224786	-1.965572

N	3.587636	-3.297033	1.920815
C	9.506374	0.770107	-3.947858
C	5.581065	2.113148	3.713686
C	3.120834	-2.624757	3.128229
H	8.520143	-0.193563	1.689439
H	6.765619	-3.801636	0.021532
H	8.656976	-1.130718	-4.406200
H	10.442259	1.597312	0.384418
H	9.284852	1.943440	1.706760
H	8.784509	1.817924	-1.185809
H	2.687970	0.705078	-2.952839
H	2.369185	2.057860	-1.814723
H	5.623066	-2.567905	-3.058433
H	5.823902	-5.123887	-1.599281
H	6.918368	-4.455686	-2.851491
H	4.199483	-4.100764	2.018110
H	10.366980	1.052322	-3.334106
H	9.772567	0.869192	-5.005758
H	8.669333	1.447398	-3.713560
H	6.371259	2.871088	3.672074
H	4.921168	2.317632	4.564373
H	6.063018	1.132136	3.865179
H	3.759833	-1.769654	3.397527
H	2.096636	-2.268100	2.987180
H	3.126846	-3.353303	3.945421
N	0.038438	2.526965	2.491922
C	0.598966	2.176185	3.798319
C	1.326818	0.817939	3.734694
H	-0.199608	2.121361	4.548687
H	1.311345	2.962889	4.084225
C	0.544213	3.699572	1.770377
C	0.714912	3.422735	0.250413
H	1.533088	3.929177	2.193370
H	-0.104548	4.579338	1.923357
N	2.250646	0.860606	2.578097
H	1.902363	0.645111	4.657010
H	0.559486	0.038179	3.636585
H	2.481173	-0.077885	2.226984
N	1.951567	3.905654	-0.396056
H	0.678249	2.333224	0.111120
H	-0.145040	3.822675	-0.293238
H	2.040799	4.919430	-0.267797
H	2.769392	3.479593	0.061402
H	4.916209	1.051085	-2.791081

H	3.787158	1.797225	2.561658
H	1.675378	1.289705	1.837397
H	2.353608	0.688929	-0.041951
C	-1.232280	1.965555	2.158754
O	-1.799370	2.517917	1.138058
O	-1.621119	0.997905	2.854082

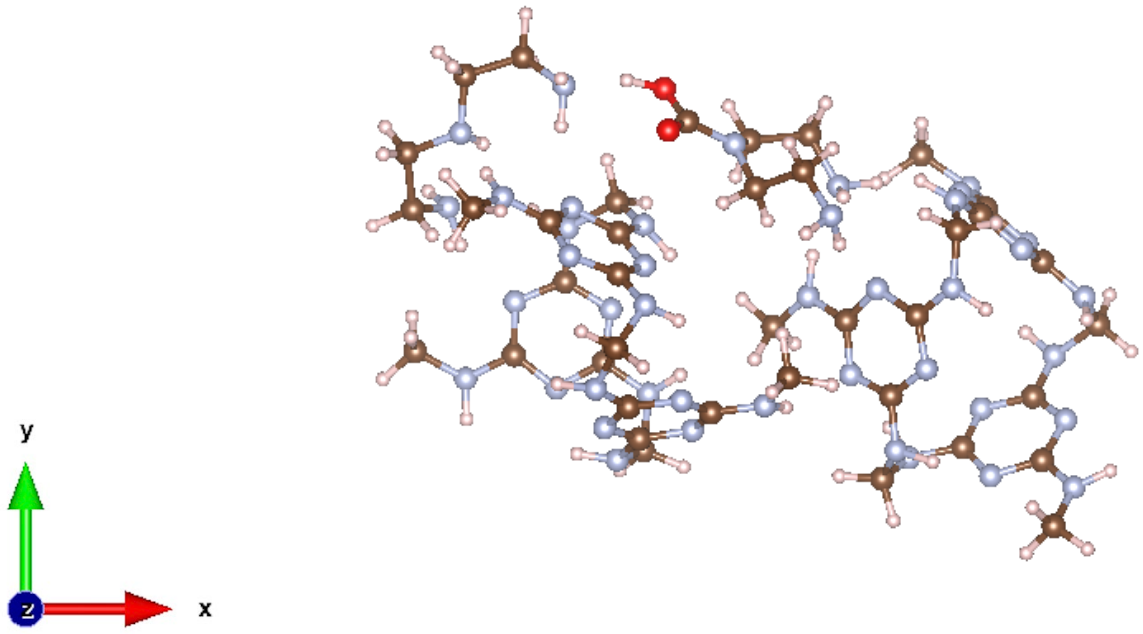

Fig. S16. DFT-calculated structure of CO_2-reacted ammonium carbamate pair within MNNs ⊃ DETA (Structure 1). The structure was obtained by DFT calculation at the TPSS-D3(BJ)/6-31G* level.

Coordinates:

	X	Y	Z
C	-0.895793	-3.049998	1.611716
N	-1.369692	-3.497857	0.417312
C	-2.702213	-3.707379	0.418905
N	-3.572525	-3.431053	1.416869
C	-2.973618	-2.911282	2.508966
N	-1.645574	-2.754833	2.700478
N	-3.800313	-2.459254	3.512951
N	-3.259981	-4.239331	-0.727527
N	0.442801	-2.872554	1.711260
C	-3.295239	-1.432118	4.447042
N	-2.557498	-0.356373	3.787145

C	-3.228822	0.423920	2.861809
N	-2.571968	0.636626	1.704800
C	-3.224184	1.464564	0.857079
N	-4.397194	2.083079	1.081790
C	-4.962312	1.748165	2.282145
N	-4.451923	0.885336	3.191361
N	-6.137898	2.353195	2.565843
N	-2.593222	1.656783	-0.346894
C	-3.261561	2.236366	-1.499806
N	-4.498120	1.580587	-1.923857
C	-4.568853	0.240130	-2.139310
N	-3.415169	-0.460825	-2.198168
C	-3.600589	-1.794388	-2.353083
N	-4.780762	-2.434949	-2.492619
C	-5.852106	-1.605771	-2.461422
N	-5.826234	-0.273669	-2.270771
N	-2.470684	-2.575039	-2.348740
C	-2.598983	-3.998674	-2.006049
N	-7.065778	-2.200983	-2.633884
C	1.056754	-2.168705	2.830315
C	-6.880127	2.085937	3.785613
C	-8.330699	-1.515584	-2.423918
H	-4.758391	-2.338048	3.190941
H	-4.267745	-4.086123	-0.763916
H	0.974809	-2.918356	0.839042
H	-2.611823	-1.905697	5.156259
H	-4.156851	-1.015042	4.970592
H	-1.715885	-0.748150	3.354017
H	-2.535045	2.209205	-2.317940
H	-3.510409	3.277678	-1.281170
H	-1.675039	-2.109695	-1.918248
H	-3.189096	-4.493296	-2.779593
H	-1.590272	-4.412717	-1.969699
H	-7.039895	-3.214793	-2.652837
H	0.631758	-2.538273	3.767667
H	2.133158	-2.363924	2.814680
H	0.887781	-1.080520	2.773154
H	-6.186399	2.007332	4.629421
H	-7.577555	2.914646	3.956301
H	-7.454055	1.146742	3.727765
H	-8.442114	-1.164622	-1.386332
H	-8.407858	-0.649100	-3.091861
H	-9.141424	-2.213708	-2.654425
N	-7.191833	3.964841	0.399190

C	-8.538445	3.459812	0.108254
C	-8.392559	2.049477	-0.485167
H	-9.079871	4.113393	-0.606404
H	-9.133728	3.425548	1.032506
C	-7.083737	5.410087	0.633603
H	-6.624496	3.693789	-0.415907
C	-5.634567	5.871135	0.397890
H	-7.393193	5.624834	1.668480
H	-7.751590	5.989832	-0.034703
N	-7.503724	2.101583	-1.661179
H	-7.906491	1.400293	0.254101
H	-9.393852	1.635591	-0.698606
H	-7.192049	1.151522	-1.927577
N	-4.592840	5.159032	1.155244
H	-1.931712	0.911910	-0.548522
H	-6.543199	2.945322	1.811682
H	-8.001068	2.496934	-2.465181
H	-5.389091	2.019693	-1.665049
H	-4.670369	5.322478	2.162990
H	-3.016018	5.282019	0.691779
H	-4.665405	4.133959	1.028876
H	-5.397765	5.744507	-0.670243
H	-5.553768	6.944057	0.615776
C	7.411401	-4.245546	-0.795614
N	8.000800	-3.229149	-1.470073
C	7.187530	-2.648265	-2.377741
N	5.890930	-2.951425	-2.614380
C	5.417501	-3.922489	-1.808114
N	6.121566	-4.635587	-0.896428
N	4.067364	-4.183630	-1.891694
N	7.716440	-1.620454	-3.125171
N	8.208101	-4.922894	0.076015
C	3.393476	-4.712694	-0.697874
N	3.742964	-3.999770	0.536385
C	3.613980	-2.626348	0.554344
N	4.505973	-1.951222	1.297401
C	4.270381	-0.612217	1.345075
N	3.269052	0.050324	0.725445
C	2.495126	-0.747200	-0.068913
N	2.603689	-2.097982	-0.176218
N	1.531263	-0.138844	-0.795685
N	5.162708	0.078215	2.111497
C	5.288450	1.530190	2.217096
N	5.230288	2.275522	0.963940

C	5.863901	1.904002	-0.187565
N	6.901388	1.049518	-0.103865
C	7.427062	0.732709	-1.312909
N	7.026679	1.194230	-2.516627
C	5.995067	2.071845	-2.443593
N	5.353055	2.454598	-1.322454
N	8.474347	-0.154817	-1.312260
C	8.819036	-0.845015	-2.558715
N	5.577482	2.600299	-3.626456
C	7.703671	-5.901014	1.028969
C	0.659982	-0.892488	-1.684663
C	4.380140	3.416993	-3.761417
H	3.569559	-3.421011	-2.348578
H	6.995046	-1.040770	-3.553322
H	9.124058	-4.510019	0.217434
H	3.680474	-5.756299	-0.552363
H	2.318081	-4.641891	-0.871375
H	4.660866	-4.278464	0.885809
H	6.250281	1.705140	2.709975
H	4.474425	1.924113	2.839143
H	8.534244	-0.714689	-0.464331
H	9.108572	-0.098572	-3.300450
H	9.650500	-1.517056	-2.343451
H	6.001451	2.189555	-4.451357
H	7.012684	-6.576353	0.515734
H	8.550304	-6.474920	1.420330
H	7.170006	-5.429001	1.869934
H	0.191769	-1.713803	-1.127217
H	-0.116247	-0.214852	-2.058245
H	1.201444	-1.323499	-2.541102
H	3.456793	2.826180	-3.652158
H	4.386629	4.198426	-2.994134
H	4.390355	3.885918	-4.750610
N	-0.314990	3.685968	0.770535
C	0.061484	3.766757	-0.642727
C	1.568311	3.915194	-0.906380
H	-0.459971	4.634109	-1.052548
H	-0.291193	2.868260	-1.180662
C	0.216398	2.581175	1.581448
C	1.410993	3.005986	2.457960
H	0.507633	1.765887	0.903161
H	-0.594335	2.207134	2.213871
N	2.294021	2.632888	-0.795508
H	1.685481	4.249896	-1.946673

H	1.975136	4.710223	-0.257628
H	3.250436	2.708002	-1.176355
N	2.117154	1.887230	3.110690
H	2.143679	3.563927	1.853004
H	1.059510	3.702798	3.228868
H	1.483796	1.439506	3.781472
H	2.326376	1.174429	2.396896
H	5.995723	-0.467612	2.307690
H	1.651671	0.886306	-0.943105
H	2.422290	2.361467	0.184016
H	4.359722	2.767832	0.789584
C	-1.499788	4.249360	1.216087
O	-1.973764	4.028069	2.336633
O	-2.073895	5.075916	0.309281

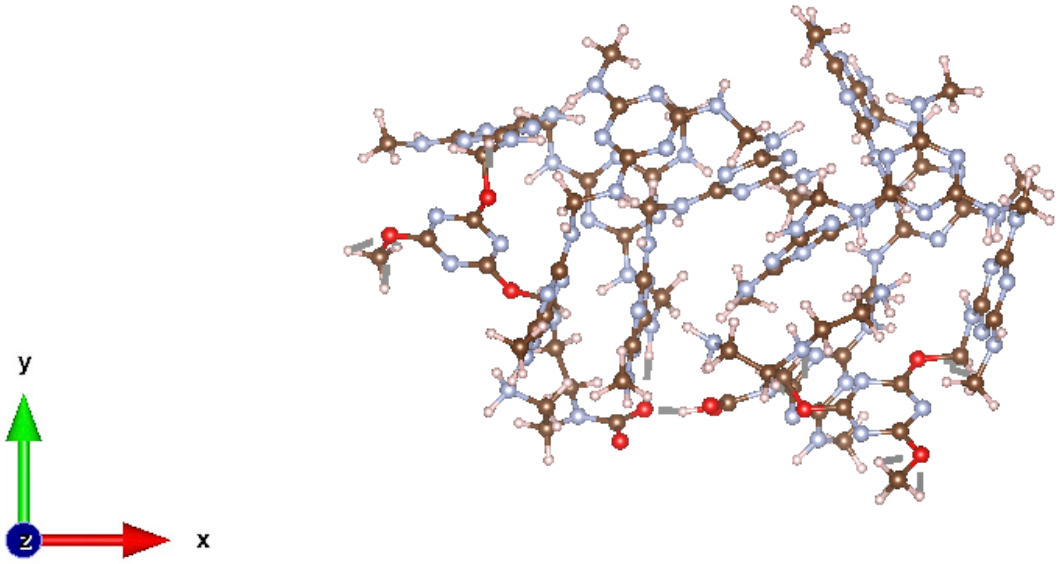

Fig. S17. DFT-calculated structure of CO_2-reacted mixed ammonium carbamate and carbamic acid within MNNs^{cya} ⊃ DETA (Structure 1). The structure was obtained by DFT calculation at the TPSS-D3(BJ)/6-31G* level.

Coordinates:

	X	Y	Z
C	3.008037	-5.411618	-4.429463
N	2.098896	-5.585046	-3.434504
C	2.102350	-4.592498	-2.540014
N	2.890308	-3.498548	-2.564541
C	3.819509	-3.508619	-3.538447
N	3.936538	-4.434638	-4.519545
N	4.729666	-2.488645	-3.567639

N	1.272389	-4.697489	-1.404127
N	2.976267	-6.369393	-5.399750
C	4.854560	-1.452235	-2.555880
C	1.803857	-4.242411	-0.157326
N	4.925918	-0.143952	-3.190559
C	5.905874	0.790862	-3.036932
N	5.559571	2.025393	-3.503331
C	6.525567	2.951281	-3.370041
N	7.737325	2.771592	-2.803567
C	7.942876	1.515975	-2.345178
N	7.080041	0.459867	-2.453494
N	9.120739	1.281156	-1.709759
N	6.298049	4.210923	-3.870880
C	10.020463	2.388287	-1.406994
C	4.960326	4.734388	-4.085613
N	4.592407	5.770920	-3.124502
C	4.348877	5.562214	-1.786704
N	4.773910	4.409378	-1.235557
C	4.545596	4.338613	0.098809
N	3.979131	5.319709	0.865709
C	3.565064	6.391414	0.158777
N	3.725627	6.591415	-1.167327
N	4.954376	3.256228	0.786802
N	2.895608	7.370596	0.840426
C	2.902082	7.460086	2.295418
C	7.833672	-3.511573	-1.117561
N	8.235997	-2.164898	-0.836550
O	6.491515	-6.894640	2.184472
C	5.776752	-6.091245	1.363673
N	6.546874	-5.299271	0.586165
C	5.804194	-4.471419	-0.163093
N	4.472423	-4.377523	-0.189838
C	3.839513	-5.271374	0.584353
N	4.445302	-6.164606	1.405704
O	2.514470	-5.342816	0.552247
O	6.387816	-3.631668	-1.044761
C	5.706396	-7.749532	3.046567
N	2.229723	0.819407	3.740269
C	2.359280	0.498497	2.408583
N	3.433171	1.004331	1.751876
C	3.401988	0.712133	0.427560
N	2.460237	0.018800	-0.235839
C	1.482050	-0.484909	0.563742
N	1.396865	-0.290034	1.915768

N	4.453309	1.145266	-0.340099
N	0.522259	-1.239585	0.008475
C	0.456606	-1.486787	-1.428233
C	8.566625	-1.770838	0.433949
N	8.564713	-2.704179	1.399292
C	8.993244	-2.259264	2.596573
N	9.434268	-1.001531	2.876419
C	9.349200	-0.157685	1.827090
N	8.913172	-0.471978	0.584072
N	9.754288	1.140670	2.019703
N	8.992163	-3.136966	3.633220
C	5.465240	2.036149	0.185973
C	8.333522	-4.439385	3.588379
N	8.909575	1.420298	4.311304
C	7.699249	2.050882	4.081042
N	7.733113	3.260376	3.476709
C	6.504786	3.821825	3.359041
N	5.331362	3.283633	3.771566
C	5.444790	2.048673	4.319172
N	6.617143	1.374479	4.496640
N	4.329949	1.410147	4.746776
N	6.419399	5.051347	2.769226
C	7.501522	5.541550	1.923293
C	2.995048	1.869201	4.378516
C	10.005594	1.634461	3.382527
C	3.863595	-6.421964	-6.544263
N	-0.606439	-3.062895	2.007476
C	-0.062601	-3.336015	3.349555
C	1.273324	-4.097270	3.241957
N	2.426931	-3.295340	2.789127
C	3.123600	-2.605537	3.879324
C	4.466484	-2.041591	3.398586
N	5.137244	-1.354762	4.526625
H	5.455327	-2.575546	-4.272334
H	2.253771	-7.069786	-5.267086
H	3.976160	-1.513788	-1.906093
H	5.751337	-1.578290	-1.949708
H	0.986399	-3.989279	0.507493
H	2.511862	-3.432635	-0.311054
H	4.076099	0.207225	-3.622439
H	9.095840	0.523769	-1.006169
H	7.005558	4.882888	-3.582244
H	10.391850	2.839453	-2.333856
H	10.861828	1.984367	-0.833422

H	9.527141	3.185575	-0.826469
H	4.894358	5.208353	-5.073219
H	4.281429	3.877372	-4.060736
H	4.123620	6.596242	-3.486110
H	4.596247	3.157518	1.741672
H	2.748540	8.217201	0.300116
H	2.543933	6.521508	2.729517
H	2.226764	8.268770	2.593376
H	3.907548	7.658457	2.700451
H	8.296163	-4.196770	-0.406374
H	8.087085	-3.752170	-2.153818
H	7.964443	-1.415544	-1.487163
H	6.446878	-8.304562	3.629393
H	5.082757	-8.426009	2.452618
H	5.058942	-7.149599	3.695699
H	1.347374	0.527959	4.147109
H	4.220889	1.222911	-1.325767
H	-0.015820	-1.817646	0.683081
H	1.408424	-1.876471	-1.808482
H	0.216409	-0.555583	-1.956307
H	-0.331911	-2.219062	-1.610202
H	9.326942	1.802674	1.376145
H	9.246412	-2.731636	4.528791
H	6.026217	1.517035	0.968979
H	6.131979	2.306434	-0.633036
H	8.148474	-4.692930	2.541913
H	7.369251	-4.410910	4.118298
H	8.974612	-5.201436	4.048958
H	8.773632	0.426403	4.503779
H	4.453053	0.385981	4.908931
H	5.471473	5.283319	2.454046
H	8.442534	5.497568	2.479311
H	7.281501	6.579776	1.650818
H	7.609808	4.945606	1.001360
H	3.117004	2.733912	3.717708
H	2.435605	2.171984	5.273833
H	10.204718	2.704074	3.304599
H	10.879439	1.110621	3.778856
H	4.536273	-5.563185	-6.470708
H	3.300352	-6.362105	-7.487828
H	4.458041	-7.348569	-6.546901
H	-0.838091	-3.941645	1.525614
H	-1.489147	-2.536119	2.066059
H	-0.760924	-3.921917	3.979768

H	0.099260	-2.368180	3.845416
H	1.143787	-4.941355	2.548334
H	1.530113	-4.529824	4.220135
H	2.073915	-2.579739	2.143433
H	3.319683	-3.351634	4.667637
H	2.547889	-1.783319	4.344065
H	4.303491	-1.290030	2.617134
H	5.062667	-2.857464	2.954325
H	5.432238	-2.042802	5.226772
H	5.987116	-0.869838	4.215264
C	-0.108358	-5.115780	-1.411205
O	-0.560994	-5.284044	-2.625084
O	-0.707688	-5.210687	-0.337713
H	-1.644100	-5.419585	-2.698007
H	-6.686864	-2.514801	-5.197609
H	-7.255269	4.810724	-4.592721
N	-6.100365	3.090984	-4.684710
H	-6.909126	2.483789	-4.574436
H	-10.686139	3.955179	-3.005390
C	-6.261507	4.479111	-4.289165
H	-5.485194	5.066112	-4.783188
H	-6.256376	-4.983718	-4.661978
N	-6.064094	-2.207690	-4.443315
C	-4.890655	2.464019	-4.435635
N	-4.871225	1.129238	-4.565155
H	-4.573351	-4.427022	-4.503781
H	-12.563510	3.225593	-1.649390
H	-11.429208	0.194299	-2.309101
H	-13.082552	0.069617	-1.598087
C	-5.504999	-4.585462	-3.953501
N	-10.568746	3.714559	-2.025485
H	-6.476269	-1.347446	-4.065843
H	-4.407506	-1.282731	-4.658018
C	-12.020326	-0.144168	-1.450769
N	-8.354172	4.143392	-2.507473
C	-11.733619	3.734337	-1.146825
N	-3.527329	-0.749749	-4.511375
C	-3.633259	0.588426	-4.352717
N	-3.865027	3.282645	-4.096328
C	-6.005305	-3.241920	-3.386334
H	-12.039914	4.762239	-0.892020
H	-1.916386	-2.059079	-4.879226
N	-6.123361	4.717145	-2.848853
C	-9.298982	3.907895	-1.563562

H	-11.850756	-1.214892	-1.295490
C	-2.692598	2.617931	-3.924259
N	-2.515786	1.283747	-4.030221
H	-6.980631	-3.385481	-2.883569
N	-5.223774	-5.578721	-2.927213
C	-2.369157	-1.510725	-4.044425
C	-7.125313	4.355591	-1.991362
O	-2.959378	-5.346123	-3.209915
H	-7.256063	-5.803071	-2.575959
H	-2.313801	5.303755	-3.694393
O	-11.660992	0.606036	-0.269589
C	-3.909178	-5.794859	-2.438710
N	-1.576779	3.342259	-3.627843
H	-11.484876	3.197885	-0.228853
H	-5.288443	-2.907486	-2.630801
H	-5.176766	4.552723	-2.503246
C	-6.309853	-5.997644	-2.044393
C	-1.674706	4.680839	-3.060826
H	-1.645931	-0.795671	-3.644433
H	-2.808641	-3.499861	-3.288977
H	-6.230470	-7.080231	-1.879737
H	1.344194	2.359211	-4.443318
N	-2.739371	-2.492196	-3.054677
N	-9.580916	-0.361975	-0.531036
N	-9.106207	3.822463	-0.232176
C	-10.379004	0.484668	0.132962
H	-0.667583	5.110298	-3.018961
H	-0.806420	2.762522	-3.258733
C	2.176884	1.884328	-3.907715
N	-6.790256	4.273484	-0.678209
H	3.077034	1.923647	-4.530924
H	1.888848	0.840420	-3.713271
O	-3.792714	-6.429712	-1.365579
O	-7.516548	-1.290003	-0.614858
C	-7.820919	3.978356	0.140952
H	-8.008604	-4.097489	-0.959449
C	-8.335433	-0.424569	-0.012234
C	-6.314323	-5.255459	-0.676736
H	-4.907278	7.198882	-0.878114
H	-2.091450	4.665727	-2.039346
C	-2.998141	-2.228708	-1.745231
N	-10.069624	1.252791	1.195945
H	-1.810536	1.054247	-1.873068
H	-5.591770	-4.435207	-0.744010

N	-7.606032	-4.685943	-0.220959
H	-3.279623	6.450803	-0.926492
N	-2.876313	-0.967569	-1.256416
C	-4.087545	6.804539	-0.267478
H	-5.655671	-2.068925	-0.691752
H	-8.284428	-5.432786	-0.044784
N	-3.366130	-3.316215	-1.043783
H	-3.712031	1.687211	-0.647070
N	2.472171	2.593602	-2.671197
N	0.371220	2.081256	-1.859908
C	-6.233074	-1.591358	0.099276
H	-5.435223	5.217973	0.177756
N	-1.696038	1.607500	-1.020073
N	-7.871904	0.311632	1.009609
H	-5.954208	-5.920402	0.113448
C	-8.781828	1.132148	1.547196
N	-7.523307	3.877845	1.478744
N	-4.631488	5.739703	0.566032
H	-9.481876	3.562994	2.075688
C	-2.785061	1.543290	-0.074192
C	1.582483	2.632581	-1.640515
H	3.429352	2.877127	-2.458596
C	-0.467241	2.128348	-0.786806
H	-5.783442	-0.665926	0.454650
H	-3.678953	7.605942	0.359135
C	-2.981876	-0.922216	0.097980
C	-8.467580	3.340932	2.412349
H	-6.932504	-3.380860	0.961305
N	-6.472742	-2.472773	1.185768
C	-3.364533	-3.151771	0.295014
H	-6.516108	3.848684	1.715411
O	-8.310482	1.891122	2.571653
N	-2.846358	0.299488	0.686872
C	-3.822332	5.025429	1.389688
H	-2.660995	2.353537	0.641526
H	-3.541001	-5.135932	0.411611
N	-3.169647	-1.970899	0.941006
N	1.982664	3.246441	-0.506139
N	-3.571964	-4.291751	0.995714
N	-0.184647	2.681022	0.415858
N	-4.393380	3.947839	2.019179
N	-2.539517	5.427289	1.503970
H	-8.265116	3.726595	3.412756
C	-6.356238	-2.118290	2.496914

C	1.052491	3.220582	0.472644
H	-5.759817	1.413278	2.564783
H	-3.055978	0.353110	1.680197
N	-5.805980	-0.917140	2.791777
C	-3.512601	3.260756	2.782093
C	-1.793847	4.652064	2.325457
N	-6.797589	-3.030559	3.393905
C	-3.707233	-4.371839	2.436540
C	-5.346200	1.737690	3.522042
H	-0.208353	5.754305	1.823214
H	-4.644479	-4.876488	2.707870
N	-3.930675	2.099639	3.369681
N	-2.216252	3.574006	3.011902
H	-5.912708	2.607999	3.884961
N	-0.483687	5.028781	2.483882
N	1.442749	3.811556	1.647772
H	-3.734119	-3.351913	2.831080
C	-5.861516	-0.613304	4.093643
H	2.323614	4.333617	1.592103
H	-2.862318	-4.913282	2.888627
N	-5.458083	0.651572	4.468574
C	-6.678609	-2.623636	4.676594
C	0.551768	4.017112	2.762579
H	-3.288443	1.721089	4.060932
H	0.023442	3.092175	3.010165
N	-6.270728	-1.409089	5.109274
H	-7.485131	-4.368915	5.241855
N	-7.016686	-3.553422	5.624672
H	-5.665020	0.868621	5.438765
H	1.166428	4.334911	3.613202
C	-7.218424	-3.213069	7.023144
H	-8.201664	-2.748872	7.211457
H	-6.442395	-2.501551	7.318415
H	-7.130958	-4.123592	7.628052

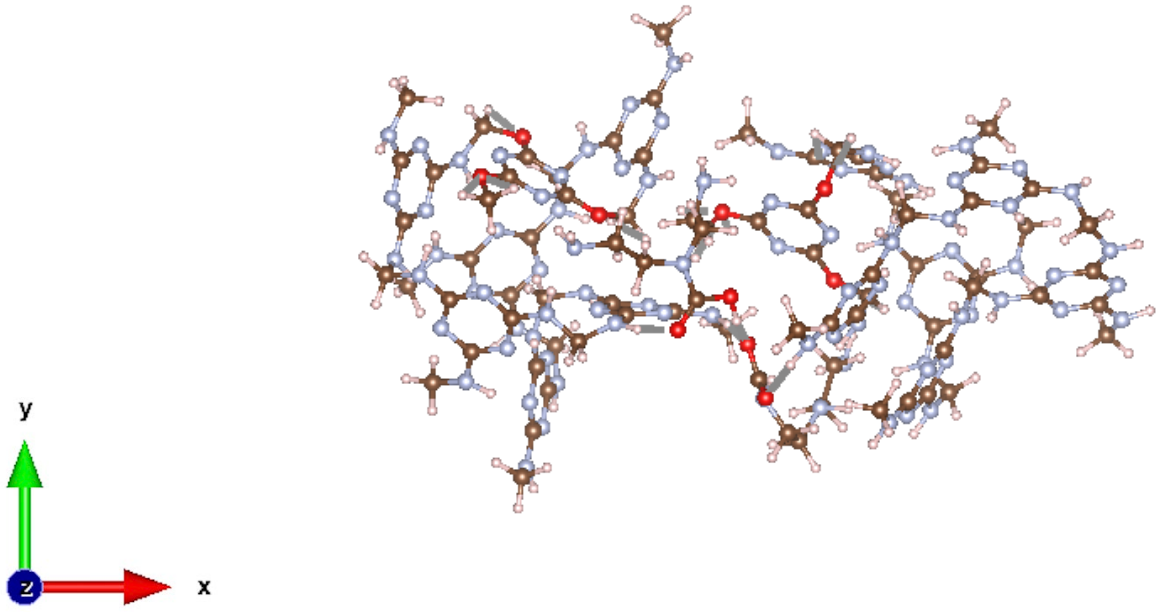

Fig. S18. DFT-calculated structure of CO_2-reacted mixed ammonium carbamate and carbamic acid within MNNs^{cya} ⊃ DETA (Structure 2). The structure was obtained by DFT calculation at the TPSS-D3(BJ)/6-31G* level.

Coordinates:

	X	Y	Z
C	-2.466849	5.869405	-1.486138
N	-2.222828	5.066479	-0.426743
C	-2.850471	3.869525	-0.506100
N	-3.605262	3.450277	-1.552520
C	-3.868433	4.403140	-2.452862
N	-3.319548	5.638845	-2.512984
N	-4.794777	4.089379	-3.429666
N	-2.703532	3.016477	0.543541
N	-1.776829	7.057438	-1.497662
C	-5.860671	3.146216	-3.130130
C	-3.424976	1.796284	0.632215
N	-5.795109	1.992331	-4.025361
C	-6.764934	1.033538	-4.040477
N	-6.442942	-0.091745	-4.732721
C	-7.458280	-0.967702	-4.830488
N	-8.695455	-0.838146	-4.301204
C	-8.860491	0.288841	-3.574059
N	-7.938061	1.287404	-3.409889
N	-10.053297	0.442718	-2.942580
N	-7.246018	-2.111635	-5.567863
C	-11.027954	-0.640217	-2.924451

C	-5.907827	-2.714662	-5.636630
N	-5.857951	-4.035579	-5.033974
C	-6.164528	-4.250488	-3.710031
N	-6.218674	-3.161701	-2.919589
C	-6.553470	-3.442961	-1.636405
N	-6.802162	-4.687143	-1.137912
C	-6.681826	-5.675777	-2.045040
N	-6.366392	-5.538222	-3.356842
N	-6.683307	-2.415252	-0.769262
N	-6.927748	-6.947521	-1.613811
C	-7.071197	-7.276157	-0.199733
C	-8.408963	4.875953	-1.155959
N	-9.217082	3.717920	-1.408303
O	-8.482384	3.149400	3.369389
C	-7.515113	3.142945	2.433132
N	-7.828477	3.874474	1.343706
C	-6.879763	3.832420	0.403413
N	-5.717731	3.168980	0.457825
C	-5.519300	2.510347	1.607747
N	-6.384650	2.461102	2.655426
O	-4.400542	1.815721	1.772409
O	-7.047016	4.514369	-0.751855
C	-8.240729	2.354491	4.559124
N	-3.847158	-1.947268	3.015392
C	-3.469081	-1.650594	1.736508
N	-4.443536	-1.471270	0.806824
C	-3.949467	-1.260280	-0.432994
N	-2.663103	-1.347531	-0.824647
C	-1.796972	-1.532030	0.213716
N	-2.141629	-1.574090	1.524483
N	-4.865728	-0.892889	-1.400462
N	-0.469608	-1.662913	-0.044781
C	0.023132	-2.089291	-1.350313
C	-10.107882	3.292974	-0.445345
N	-10.595144	4.225462	0.392760
C	-11.266606	3.706384	1.436062
N	-11.569715	2.400864	1.628399
C	-11.154610	1.595891	0.630576
N	-10.404042	1.974961	-0.434687
N	-11.487514	0.266466	0.728849
N	-11.687763	4.570882	2.406539
C	-6.284970	-1.056612	-1.123800
C	-11.044046	5.874775	2.566418
N	-10.631455	-0.271504	2.960605

C	-9.576387	-1.114628	2.648348
N	-9.873343	-2.110163	1.780921
C	-8.821698	-2.925819	1.527614
N	-7.580178	-2.802909	2.044949
C	-7.419640	-1.769012	2.914943
N	-8.396871	-0.867190	3.243026
N	-6.215748	-1.617101	3.502307
N	-9.017304	-3.967996	0.670729
C	-10.169328	-3.997461	-0.219407
C	-5.121171	-2.567875	3.318552
C	-11.785811	-0.260612	2.069795
C	-2.165848	8.167731	-2.355939
N	-0.757504	2.986362	2.639375
C	-1.111334	1.633745	3.122518
C	-0.823807	1.345786	4.612836
N	-1.485468	0.114772	5.070475
C	-2.832747	0.195312	5.642375
C	-3.834411	0.850018	4.679626
N	-5.247055	0.679712	5.092084
H	-5.017732	4.875301	-4.033927
H	-1.956874	3.206346	1.261390
H	-1.340524	7.269113	-0.604696
H	-5.757458	2.847641	-2.082712
H	-6.837325	3.617433	-3.280581
H	-2.783628	0.965648	0.921502
H	-3.972669	1.601878	-0.289137
H	-4.865102	1.663265	-4.271088
H	-10.077487	1.075326	-2.129602
H	-8.013633	-2.772892	-5.464306
H	-11.368213	-0.864307	-3.941989
H	-11.878294	-0.310650	-2.317338
H	-10.609696	-1.566397	-2.500105
H	-5.585339	-2.829809	-6.678484
H	-5.230884	-2.027661	-5.120873
H	-5.973862	-4.852113	-5.625994
H	-6.644133	-2.647111	0.232892
H	-6.660672	-7.672072	-2.271016
H	-6.198471	-6.956600	0.387491
H	-7.194565	-8.360803	-0.113526
H	-7.958250	-6.784202	0.217110
H	-8.899015	5.474014	-0.385652
H	-8.226763	5.444287	-2.070698
H	-8.814038	2.986089	-2.010188
H	-9.136854	2.505973	5.167952

H	-7.356246	2.729598	5.086929
H	-8.119489	1.295970	4.294294
H	-3.059235	-2.152195	3.652023
H	-4.581242	-1.110458	-2.353149
H	0.067219	-1.978315	0.792613
H	0.252586	-3.167176	-1.341935
H	0.925110	-1.522959	-1.608161
H	-0.754664	-1.903855	-2.097404
H	-10.897226	-0.354309	0.174500
H	-11.992238	4.091216	3.248890
H	-6.558906	-0.427140	-0.274309
H	-6.830521	-0.743082	-2.013397
H	-9.964472	5.767807	2.743975
H	-11.516013	6.387865	3.411425
H	-11.192567	6.466841	1.658346
H	-10.341189	0.644424	3.297977
H	-6.057114	-0.837419	4.167011
H	-8.152647	-4.376688	0.286563
H	-11.088435	-3.870027	0.360753
H	-10.184402	-4.968617	-0.726472
H	-10.123431	-3.201697	-0.981460
H	-5.434821	-3.253237	2.526585
H	-4.969617	-3.127873	4.250887
H	-12.132789	-1.286382	1.946879
H	-12.556368	0.368479	2.515280
H	-2.294392	7.796131	-3.376542
H	-1.368262	8.919379	-2.338714
H	-3.113030	8.636881	-2.041000
H	-0.825506	3.674164	3.394833
H	0.199538	2.992913	2.265052
H	-0.586408	0.894475	2.510016
H	-2.182428	1.486486	2.962341
H	-1.212949	2.170013	5.233161
H	0.254327	1.260471	4.789401
H	-2.788596	0.755814	6.593181
H	-3.133584	-0.834450	5.857157
H	-3.743704	0.355752	3.710713
H	-3.576898	1.915082	4.542077
H	-5.386715	1.063084	6.034274
H	-5.792121	1.274899	4.452370
C	-1.023826	-1.120987	4.624646
O	0.160441	-1.021803	4.035461
O	-1.664058	-2.175178	4.809375
H	0.374428	-1.860276	3.407788

H	2.074043	-1.153366	6.041735
H	5.349045	1.214318	4.916310
H	13.292890	1.827432	3.017133
C	2.501901	-2.162829	6.098235
H	3.381965	-2.134937	6.755392
H	8.210347	1.249272	3.841198
H	1.753859	-2.845917	6.512656
N	5.756562	0.867032	4.053038
H	6.032786	2.848790	3.470361
C	12.591912	1.481081	2.248185
H	11.762295	0.945425	2.716397
N	7.740996	1.677415	3.049093
N	4.260478	-0.839608	4.451158
C	6.289197	1.835836	3.123696
N	9.869964	2.130908	2.340604
N	11.994420	2.621837	1.546974
C	5.079954	-0.246531	3.573479
C	8.536898	2.303673	2.146212
N	2.854563	-2.655449	4.775197
H	5.654370	-5.716035	4.449353
N	13.337154	0.587985	1.375575
C	10.628022	2.722866	1.403461
H	4.348442	4.471422	2.621667
H	14.317727	0.788589	1.199408
H	5.908320	-4.028697	3.911755
C	5.351050	-4.948622	3.723503
C	3.639525	-1.944067	3.948164
H	4.277235	-4.749064	3.815169
H	12.501361	2.981974	0.741151
H	5.865356	1.664846	2.130895
N	7.950487	3.010259	1.154026
O	3.436146	2.729858	2.153648
C	3.932993	4.041573	1.709114
H	2.244051	-3.382547	4.333887
N	10.198627	3.446219	0.345096
N	5.324929	-0.600070	2.298165
H	7.886040	-1.349972	2.101294
C	12.741843	-0.134227	0.367178
C	8.849458	3.540671	0.268019
N	11.404571	-0.280226	0.419760
H	3.091112	4.624168	1.326982
H	5.883892	3.611592	0.943589
N	3.737188	-2.372285	2.652838
N	7.004544	-3.556511	1.845996

N	4.944803	3.967312	0.699671
N	5.670266	-5.393292	2.369106
N	8.189936	-1.700273	1.194940
H	9.030425	0.140896	0.801037
C	4.538526	-1.626662	1.893712
C	2.855179	2.007271	1.146872
N	13.592222	-0.618649	-0.567186
O	1.383754	-4.529453	3.432215
C	6.333708	-4.652812	1.444703
C	7.577882	-2.867418	0.836920
C	8.588393	-0.675289	0.229725
N	8.325460	4.217850	-0.778672
N	1.573924	2.257139	0.872992
H	5.114699	-6.143932	1.970497
H	9.692399	5.692610	-1.429038
C	10.900356	-0.971518	-0.628924
N	3.649406	1.123743	0.533413
H	3.057490	-5.604017	1.739291
C	9.154036	4.815216	-1.813703
C	4.607676	3.829666	-0.625960
H	7.326325	4.035876	-0.979849
C	1.146754	-3.891934	2.368255
N	9.563339	-1.136018	-0.732234
C	12.953854	-1.289620	-1.555274
N	4.583130	-1.952457	0.541703
O	0.755474	-2.666343	2.263800
H	7.721610	-0.308922	-0.335474
H	5.499105	-0.194563	0.016374
H	9.901437	4.108307	-2.198642
C	2.154543	-5.746236	1.122004
N	3.330829	4.055459	-0.986732
H	1.606566	-6.599097	1.543999
C	1.124017	1.595128	-0.221459
N	6.274486	-5.130004	0.165752
N	11.624169	-1.481058	-1.667832
N	7.636265	-3.238150	-0.472802
N	5.620154	3.497802	-1.455233
C	4.947809	-1.007987	-0.454974
C	3.057989	0.488796	-0.493452
N	1.314091	-4.555557	1.138939
H	14.711313	-1.504807	-2.503427
H	3.965213	-2.720945	0.270317
N	13.746792	-1.819279	-2.534175
H	0.852350	4.390017	-0.936687

O	-0.125688	1.914737	-0.556727
H	8.492698	5.127968	-2.629018
C	7.008435	-4.409015	-0.709401
H	10.179429	1.717893	-2.324335
H	9.220553	-1.734463	-1.488610
N	1.819210	0.719328	-0.963301
O	3.746409	-0.453914	-1.152168
H	1.585499	-3.912987	0.393768
C	2.544459	-6.029509	-0.330735
H	4.244689	-4.913651	-0.505302
H	3.117910	-6.973957	-0.401129
H	5.490013	-1.464603	-1.282416
C	0.744368	4.640457	-1.993107
C	3.086440	3.825899	-2.296766
C	10.399018	1.429075	-3.364965
C	5.238581	3.291174	-2.738311
N	3.281808	-4.873131	-0.872597
H	0.746255	5.734242	-2.109152
H	11.473115	1.553970	-3.545038
C	-0.690852	1.324111	-1.759237
H	10.619946	-0.670486	-3.154980
C	13.206699	-2.349919	-3.778473
N	7.108950	-4.959877	-1.963330
H	-1.053569	0.317976	-1.538071
H	7.092007	2.602622	-3.173625
N	10.040354	0.040425	-3.624667
N	1.823927	4.014466	-2.751358
H	-0.211560	4.251085	-2.352537
N	3.988408	3.392998	-3.221167
H	-1.532098	1.974708	-2.009196
N	6.252556	2.983601	-3.611902
H	12.319994	-2.946559	-3.546147
H	9.838356	2.077881	-4.043414
H	1.627925	-6.150204	-0.926299
H	8.662790	-3.832407	-2.791384
C	8.752718	-0.349243	-3.820368
H	6.526540	-5.778939	-2.105374
H	13.964953	-2.984832	-4.250694
N	8.500842	-1.664925	-3.612542
H	4.662817	-3.135590	-2.218276
C	7.746358	-4.335394	-3.107405
H	12.909692	-1.556307	-4.484084
H	0.061548	1.304859	-2.553069
C	3.347681	-4.822397	-2.336595

N	7.847318	0.582144	-4.216121
C	3.750560	-3.409798	-2.762553
H	2.966337	-2.701514	-2.457490
C	7.227987	-2.036051	-3.874213
H	1.721541	3.891137	-3.753752
C	6.610870	0.074493	-4.426961
N	6.904502	-3.348657	-3.764329
H	4.051891	-5.557479	-2.778117
C	5.956061	2.385709	-4.907565
H	7.996277	-5.142895	-3.807847
N	6.222243	-1.205827	-4.272448
H	2.348933	-5.062241	-2.732716
N	5.652603	0.960398	-4.862723
H	5.938519	-3.610254	-4.045299
H	6.837513	2.519867	-5.538708
H	4.693683	0.686724	-4.661666
H	5.092993	2.902612	-5.328711
N	4.092226	-3.353150	-4.197816
H	4.373426	-2.389576	-4.430949
H	3.272794	-3.586603	-4.767069

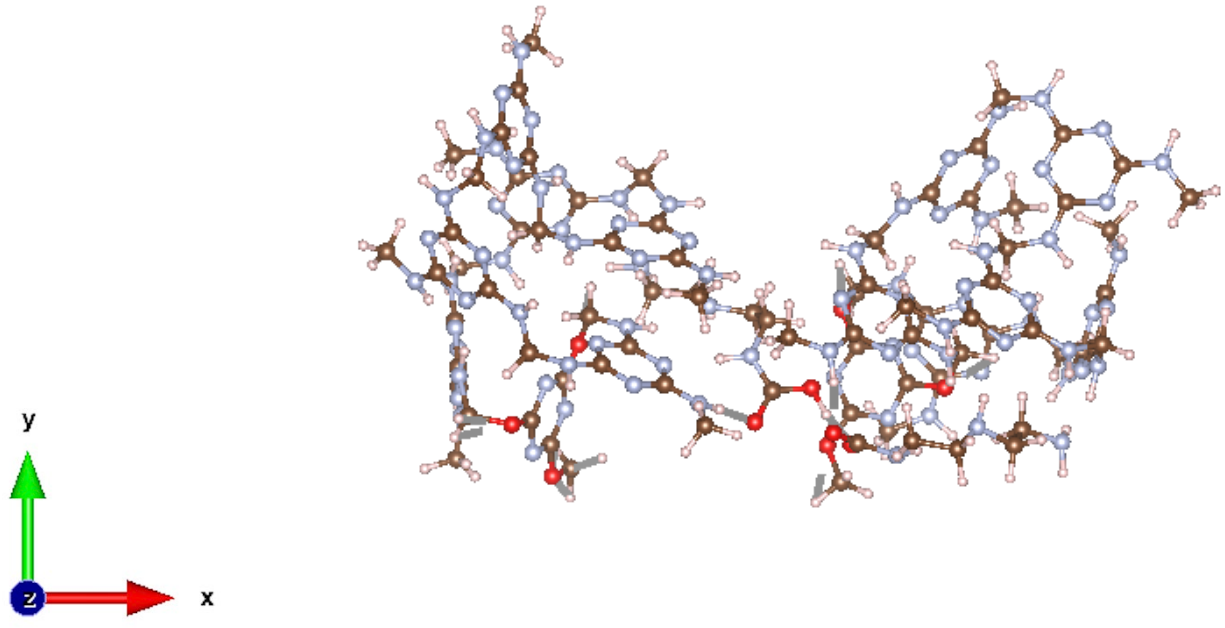

Fig. S19. DFT-calculated structure of CO_2-reacted mixed ammonium carbamate and carbamic acid within MNNs^{cya} ⊃ DETA (Structure 3). The structure was obtained by DFT calculation at the TPSS-D3(BJ)/6-31G* level.

Coordinates:

	X	Y	Z
C	-3.496614	-2.869151	2.513130
N	-3.524670	-2.241746	1.294148
C	-4.671516	-1.605521	1.030285
N	-5.746704	-1.515024	1.841129
C	-5.602429	-2.202069	2.983004
N	-4.526009	-2.887010	3.405818
N	-6.698079	-2.213710	3.830228
N	-4.753616	-0.952452	-0.185423
N	-2.355831	-3.506230	2.824281
C	-8.001326	-1.812513	3.356153
C	-6.000704	-0.597951	-0.762957
N	-8.308642	-0.472262	3.872937
C	-9.363773	0.301376	3.492598
N	-9.550240	1.412216	4.251262
C	-10.554453	2.195337	3.822447
N	-11.347434	1.981873	2.748656
C	-11.062152	0.842625	2.081171
N	-10.091787	-0.067752	2.413760
N	-11.798264	0.568579	0.976359
N	-10.839241	3.323577	4.556419
C	-12.751015	1.538352	0.454326
C	-9.778613	4.018544	5.294241
N	-9.466717	5.324378	4.741525
C	-8.937321	5.511731	3.488829
N	-8.522911	4.412663	2.828706
C	-8.019509	4.683664	1.602372
N	-7.924966	5.920798	1.034376
C	-8.362382	6.920460	1.826569
N	-8.890992	6.796208	3.067471
N	-7.617443	3.659907	0.816302
N	-8.263022	8.194034	1.343064
C	-7.906877	8.491376	-0.036792
C	-10.024090	-3.901832	1.106003
N	-10.337822	-2.583419	0.643226
O	-7.241023	-5.919037	-2.842045
C	-7.304242	-4.864230	-2.006436
N	-7.884184	-5.138829	-0.819713
C	-7.945116	-4.062103	-0.028608
N	-7.440419	-2.845039	-0.273817
C	-6.884256	-2.722657	-1.491262
N	-6.811282	-3.690486	-2.425476
O	-6.364296	-1.546463	-1.861185

O	-8.586964	-4.196340	1.171619
C	-6.621997	-5.661611	-4.123758
N	-3.353050	3.109787	-1.162398
C	-3.713920	2.465787	0.000451
N	-5.021774	2.462448	0.333111
C	-5.250691	1.866828	1.537909
N	-4.329281	1.323720	2.352701
C	-3.073973	1.335140	1.857433
N	-2.693094	1.889104	0.669301
N	-6.533786	1.835059	1.988285
N	-2.096467	0.788379	2.620675
C	-2.364187	0.045730	3.847068
C	-10.375921	-2.297986	-0.697851
N	-10.357584	-3.327150	-1.567478
C	-10.340151	-2.949189	-2.862821
N	-10.363444	-1.675512	-3.334152
C	-10.469235	-0.750470	-2.361152
N	-10.463947	-0.990220	-1.026811
N	-10.640553	0.557342	-2.731439
N	-10.274766	-3.929189	-3.803942
C	-7.668002	2.262932	1.188847
C	-10.434665	-5.347834	-3.508819
N	-8.911480	1.274845	-4.316204
C	-8.278674	2.256024	-3.583134
N	-9.079144	3.091121	-2.881434
C	-8.394640	4.036944	-2.189984
N	-7.044795	4.143044	-2.111142
C	-6.367968	3.251682	-2.873441
N	-6.934970	2.278384	-3.644080
N	-5.014451	3.321927	-2.908998
N	-9.101848	4.977018	-1.506817
C	-10.498686	4.771823	-1.147981
C	-4.219769	4.012929	-1.905930
C	-10.327480	1.015134	-4.081414
C	-2.153895	-4.238039	4.064288
N	-0.634321	-1.921723	0.378289
C	-0.087638	-0.727443	-0.238709
C	-0.569005	-0.503737	-1.672288
N	-2.027343	-0.260884	-1.738033
C	-2.391200	0.174018	-3.094292
C	-3.907880	0.229098	-3.255987
N	-4.284767	1.026362	-4.441499
H	-6.632925	-2.908583	4.567184
H	-3.905100	-0.991176	-0.767860

H	-1.678831	-3.697376	2.045221
H	-7.992672	-1.774043	2.264146
H	-8.753730	-2.538729	3.698386
H	-5.963064	0.354634	-1.288485
H	-6.770649	-0.623411	0.007770
H	-7.889041	-0.215896	4.762842
H	-11.345879	-0.055493	0.285846
H	-11.517957	3.921792	4.089135
H	-13.525490	1.748493	1.200537
H	-13.210321	1.103713	-0.440262
H	-12.272857	2.494279	0.189230
H	-10.078301	4.181550	6.336405
H	-8.903056	3.363442	5.261250
H	-9.872544	6.156315	5.157063
H	-7.073395	3.891676	-0.020949
H	-8.750217	8.891118	1.896105
H	-7.061106	7.861309	-0.327665
H	-7.616808	9.545824	-0.104804
H	-8.734551	8.300468	-0.740011
H	-10.498964	-4.635050	0.450020
H	-10.348454	-4.011495	2.141820
H	-10.163830	-1.777163	1.269659
H	-6.651600	-6.624989	-4.639624
H	-5.591217	-5.317413	-3.989442
H	-7.187081	-4.899677	-4.672604
H	-2.350455	3.160166	-1.311875
H	-6.706352	1.245003	2.797450
H	-1.180068	0.706426	2.189537
H	-3.423383	0.168364	4.080887
H	-1.752402	0.440537	4.669052
H	-2.143211	-1.017488	3.706256
H	-10.456454	1.246995	-2.004086
H	-10.364147	-3.598058	-4.759247
H	-7.728511	1.694505	0.251940
H	-8.564633	2.089671	1.782480
H	-10.105952	-5.519352	-2.481265
H	-9.809495	-5.935861	-4.189045
H	-11.484020	-5.668051	-3.610487
H	-8.324228	0.470530	-4.525833
H	-4.540941	2.656256	-3.553007
H	-8.540203	5.510268	-0.826127
H	-11.030012	4.347347	-2.003865
H	-10.940302	5.739208	-0.882692
H	-10.600590	4.089780	-0.287384

H	-4.914649	4.522112	-1.232891
H	-3.566351	4.750800	-2.388427
H	-10.871016	1.947629	-4.246191
H	-10.653632	0.245324	-4.781757
H	-2.364061	-3.593535	4.927712
H	-1.104608	-4.552018	4.079577
H	-2.805908	-5.122465	4.132578
H	0.999720	-0.820858	-0.225238
H	-0.350360	0.160711	0.361228
H	-0.325748	-1.383162	-2.281580
H	-0.006802	0.352632	-2.095532
H	-2.235517	0.507055	-1.082778
H	-1.961368	-0.553021	-3.800059
H	-1.962046	1.164165	-3.355338
H	-4.321934	0.762787	-2.391812
H	-4.325058	-0.789820	-3.251382
H	-3.896200	0.605898	-5.291251
H	-5.310373	1.029619	-4.529936
C	0.079706	-3.083903	0.574109
O	1.383356	-2.983451	0.374475
O	-0.497305	-4.128914	0.954381
H	-1.655124	-2.026005	0.425958
H	1.878246	-3.876916	0.795677
H	0.639008	-0.226868	4.019840
H	1.851836	1.763660	0.563953
H	7.379749	7.721411	-0.055802
C	0.625470	-1.292221	3.760718
H	0.188680	-1.861453	4.587885
H	4.192723	3.673008	0.225505
H	0.007968	-1.434683	2.862008
N	2.845936	1.578655	0.465006
H	2.710277	2.228718	-1.507426
C	7.698700	6.675440	-0.141075
H	7.025097	6.038089	0.438637
N	4.414412	3.041833	-0.539187
N	2.377637	-0.026931	2.058754
C	3.485203	1.935595	-0.784235
N	5.970803	4.646593	-1.011149
N	7.596834	6.220528	-1.529481
C	3.253829	0.450594	1.158611
C	5.362658	3.495946	-1.401055
N	1.986910	-1.766446	3.563888
H	4.502530	0.288740	6.281098
N	9.049907	6.611516	0.390348

C	6.950457	5.044416	-1.838106
H	2.329689	1.113632	-4.025757
H	9.639275	7.436533	0.336777
H	4.417918	0.429591	4.500961
C	4.527383	-0.269802	5.332990
C	2.790690	-1.180952	2.648381
H	3.703497	-0.991665	5.302700
H	8.406841	6.386712	-2.123229
H	4.043201	1.079753	-1.173399
N	5.629451	2.788357	-2.522418
O	2.396781	-0.374231	-2.674567
C	2.876597	0.172971	-3.937771
H	2.128789	-2.798863	3.654047
N	7.359239	4.431539	-2.971382
N	4.464718	-0.041717	0.847185
H	6.151798	1.763011	1.458576
C	9.752704	5.441564	0.549315
C	6.665404	3.305676	-3.255216
N	9.067254	4.286237	0.451043
H	2.615610	-0.520609	-4.741410
H	4.668398	1.241402	-3.464947
N	3.966887	-1.821286	2.386000
N	6.475886	0.388287	3.393422
N	4.282337	0.438001	-3.992096
N	5.806221	-0.947716	5.169707
N	7.144986	1.542591	1.526199
H	7.454892	2.496283	-0.276028
C	4.722450	-1.217710	1.467115
C	2.898310	-1.609559	-2.345163
N	11.073344	5.603910	0.798451
O	2.110850	-4.488033	3.421887
C	6.715508	-0.657266	4.206459
C	7.413880	0.564334	2.439665
C	7.899648	1.672626	0.280829
N	7.017079	2.615961	-4.366934
N	2.136594	-2.661109	-2.632386
H	5.997844	-1.791178	5.699477
H	8.012141	4.022834	-5.574655
C	9.845265	3.189993	0.606061
N	4.089026	-1.602875	-1.728483
H	4.550903	-3.654109	4.270283
C	8.161609	3.013874	-5.172347
C	5.181712	-0.540569	-4.317716
H	6.766284	1.609141	-4.363225

C	2.921823	-4.824459	2.511365
N	9.300622	1.959151	0.489192
C	11.728991	4.426430	0.925533
N	5.908362	-1.858301	1.120902
O	2.707570	-4.871500	1.237545
H	7.847180	0.749023	-0.311129
H	6.049578	-0.887596	-0.699424
H	9.099724	3.019307	-4.596692
C	4.733932	-4.736869	4.158348
N	4.715078	-1.739633	-4.716345
H	4.228650	-5.251770	4.986214
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N	8.598964	-0.096646	2.336054
N	6.484646	-0.187657	-4.239970
C	6.362460	-1.824201	-0.236037
C	4.566705	-2.835809	-1.490515
N	4.217272	-5.230465	2.885485
H	13.450641	5.446578	1.113858
H	6.014861	-2.762461	1.598582
N	13.065722	4.511422	1.195601
H	3.288755	-3.580240	-4.954260
O	1.905141	-4.892598	-2.363961
H	8.249436	2.299682	-5.998538
C	8.736206	-1.064572	3.264577
H	10.293894	2.714395	-2.595391
H	9.892619	1.164971	0.748825
N	3.932546	-3.992005	-1.731598
O	5.811248	-2.972182	-0.989607
H	4.880146	-5.065368	2.126553
C	6.238407	-5.017342	4.194471
H	7.125527	-3.337515	3.433553
H	6.661501	-4.760594	5.184231
H	7.438575	-1.989574	-0.302513
C	3.934689	-4.401379	-5.272748
C	5.682674	-2.651671	-4.950342
C	11.297358	2.264540	-2.673914
C	7.349737	-1.176666	-4.561022
N	6.902697	-4.285173	3.096852
H	3.627100	-4.743261	-6.271421
H	12.037465	3.070482	-2.745276
C	2.361066	-6.110458	-1.702687
H	11.713518	1.933745	-0.618371

C	13.955390	3.359890	1.186023
N	9.936868	-1.728327	3.330926
H	2.580048	-5.900342	-0.649227
H	8.910127	-0.000958	-4.024391
N	11.612598	1.438117	-1.516762
N	5.305849	-3.909400	-5.295527
H	3.833926	-5.229882	-4.560420
N	7.027264	-2.443112	-4.876857
H	1.517504	-6.796659	-1.810554
N	8.675462	-0.815581	-4.591353
H	13.403956	2.498809	1.573870
H	11.343422	1.646757	-3.574800
H	6.399906	-6.092802	4.031946
H	11.160068	-0.475864	2.187252
C	11.162364	0.162028	-1.388330
H	9.952697	-2.503013	3.986094
H	14.817245	3.566848	1.831494
N	11.202672	-0.347371	-0.132656
H	8.390373	-3.275091	1.198382
C	11.053117	-1.536653	2.424437
H	14.317081	3.109962	0.174465
H	3.251470	-6.501302	-2.210399
C	8.129282	-4.896988	2.578734
N	10.738468	-0.493041	-2.499304
C	8.379521	-4.372474	1.162768
H	7.537739	-4.671905	0.517303
C	10.813745	-1.638012	-0.039461
H	6.074760	-4.566662	-5.374239
C	10.365410	-1.771177	-2.259570
N	10.905092	-2.253607	1.166281
H	9.026477	-4.711126	3.203694
C	9.720146	-1.829928	-4.621747
H	11.950615	-1.886448	2.950779
N	10.363241	-2.407761	-1.072121
H	7.983102	-5.987654	2.549313
N	9.958020	-2.504454	-3.349741
H	10.654791	-3.261978	1.186329
H	10.646345	-1.332579	-4.918408
H	9.364175	-3.300480	-3.129701
H	9.435941	-2.587371	-5.353359
N	9.706145	-4.785974	0.662718
H	9.838031	-4.393120	-0.279986
H	9.754499	-5.807108	0.588899

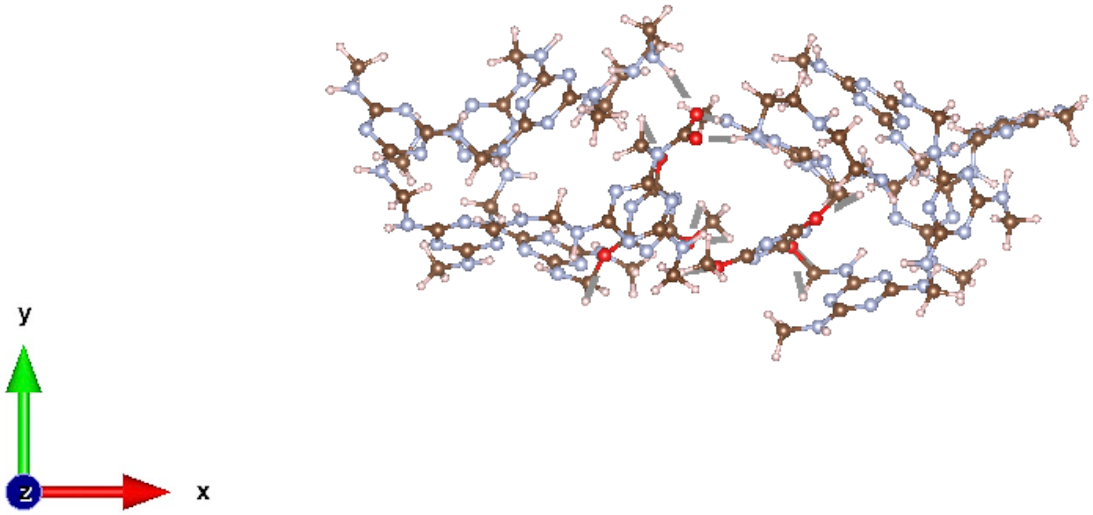

Fig. S20. DFT-calculated structure of CO_2-reacted ammonium carbamate pair within MNNs^{cya} ⊃ DETA (Structure 1). The structure was obtained by DFT calculation at the TPSS-D3(BJ)/6-31G* level.

Coordinates:

	X	Y	Z
C	1.904439	1.739572	-0.749865
N	2.645498	1.462409	0.379736
C	3.864833	0.953674	0.130437
N	4.394219	0.697847	-1.075793
C	3.534728	0.913545	-2.089559
N	2.304245	1.452144	-2.014986
N	3.960924	0.538587	-3.341211
N	4.678737	0.639303	1.220783
N	0.700448	2.304144	-0.581828
C	5.040483	-0.414377	-3.498603
C	5.296147	-0.653186	1.240080
N	6.297319	0.289408	-3.777359
C	7.479034	-0.397607	-3.930385
N	8.461085	0.293712	-4.548420
C	9.632309	-0.370969	-4.605355
N	9.888772	-1.602786	-4.111105
C	8.823929	-2.184001	-3.517098
N	7.563104	-1.646430	-3.423897
N	8.999010	-3.399168	-2.951927
N	10.676393	0.245790	-5.246356
C	10.300039	-4.052266	-2.927021
C	10.781790	1.708372	-5.278885

N	11.866384	2.211085	-4.456754
C	11.908090	2.093896	-3.090602
N	10.789332	1.655514	-2.473944
C	10.924449	1.574543	-1.131386
N	12.052522	1.858935	-0.420034
C	13.077809	2.308264	-1.175263
N	13.080632	2.450447	-2.521714
N	9.892205	1.122551	-0.381285
N	14.225923	2.657706	-0.531616
C	14.445507	2.462405	0.893766
C	4.251532	-3.857692	-3.036733
N	5.562249	-3.752739	-2.489454
O	0.319966	-3.627268	0.647690
C	1.527588	-3.166278	0.314391
N	1.799335	-3.260172	-1.006516
C	2.986195	-2.748247	-1.327328
N	3.863102	-2.153645	-0.501667
C	3.486432	-2.175773	0.786619
N	2.336350	-2.677669	1.273326
O	4.317314	-1.663716	1.718811
O	3.319145	-2.782383	-2.650859
C	-0.080151	-3.466981	2.035367
N	8.078686	3.106880	3.391265
C	7.382253	3.040704	2.213315
N	7.858637	2.250306	1.235735
C	7.164494	2.362783	0.068157
N	6.102926	3.176801	-0.153231
C	5.664522	3.803273	0.946505
N	6.245848	3.783474	2.179570
N	7.588782	1.631958	-0.988771
N	4.497775	4.515781	0.858552
C	3.804390	4.744153	-0.406188
C	5.874612	-4.255158	-1.252089
N	4.933236	-4.946044	-0.582471
C	5.326161	-5.351495	0.645387
N	6.549486	-5.155082	1.208090
C	7.414965	-4.520765	0.396817
N	7.136959	-4.020993	-0.832979
N	8.711660	-4.379524	0.812669
N	4.431564	-6.023986	1.414277
C	8.639152	0.619684	-0.884229
C	3.018795	-6.189037	1.099364
N	8.812642	-3.476454	3.091978
C	9.379519	-2.246648	2.865192

N	10.343032	-2.197341	1.918807
C	10.876798	-0.959392	1.753196
N	10.482184	0.165394	2.403102
C	9.520578	-0.029766	3.328808
N	8.915227	-1.219644	3.609034
N	9.122921	1.035490	4.076581
N	11.894449	-0.800092	0.873012
C	12.220019	-1.784905	-0.147816
C	9.329675	2.415689	3.675650
C	9.102342	-4.594141	2.198904
C	-0.209742	2.507471	-1.706963
N	1.945284	1.694327	3.112498
C	2.378646	3.114690	3.294135
C	3.412293	3.140922	4.424836
N	4.462238	2.174246	4.061604
C	5.264558	1.627777	5.165850
C	5.956635	0.353634	4.670610
N	6.985521	-0.098563	5.619812
H	3.229878	0.564921	-4.045219
H	4.331989	0.957702	2.130405
H	0.288499	2.336244	0.366070
H	5.162893	-0.987534	-2.577860
H	4.780661	-1.097096	-4.320674
H	6.093619	-0.698025	1.980353
H	5.632554	-0.919468	0.238532
H	6.224977	1.105445	-4.382612
H	8.306589	-3.667577	-2.229905
H	11.551656	-0.263993	-5.141484
H	10.648348	-4.250301	-3.947554
H	10.184818	-5.000310	-2.390986
H	11.061835	-3.439207	-2.424032
H	10.971100	2.054030	-6.301365
H	9.819622	2.093927	-4.929960
H	12.755153	2.445055	-4.887515
H	9.984409	1.206456	0.635222
H	15.006607	2.878983	-1.140175
H	13.498693	2.625798	1.416448
H	15.188621	3.188331	1.242601
H	14.800286	1.445865	1.129949
H	3.807283	-4.810202	-2.739401
H	4.299502	-3.753282	-4.121728
H	6.205534	-3.026845	-2.853904
H	-1.108289	-3.831509	2.064123
H	-0.033145	-2.413942	2.326235

H	0.566497	-4.063388	2.687289
H	7.690703	3.740798	4.081468
H	6.942513	1.558308	-1.775078
H	4.357510	5.179686	1.614315
H	3.988623	3.880289	-1.048413
H	4.167703	5.647528	-0.919519
H	2.728711	4.834368	-0.216697
H	9.252231	-3.662794	0.330644
H	4.761498	-6.226835	2.352647
H	8.320172	-0.190319	-0.213166
H	8.803646	0.227442	-1.886376
H	2.872799	-5.920347	0.050858
H	2.396416	-5.533333	1.724152
H	2.716308	-7.232195	1.256554
H	7.908656	-3.460829	3.555504
H	8.440679	0.834414	4.828769
H	12.122887	0.182831	0.660868
H	12.064199	-2.786327	0.261877
H	13.271154	-1.665796	-0.434481
H	11.593145	-1.658487	-1.045721
H	9.973527	2.411714	2.792825
H	9.818579	2.975790	4.481907
H	10.180848	-4.767380	2.210080
H	8.566954	-5.468547	2.570728
H	0.241999	3.189879	-2.438162
H	-1.128589	2.932584	-1.297315
H	-0.439019	1.552226	-2.194625
H	2.681374	1.098899	3.526238
H	0.978918	1.549978	3.573594
H	1.482323	3.706620	3.497540
H	2.833714	3.441123	2.350969
H	2.945098	2.817095	5.365943
H	3.792188	4.165765	4.571466
H	5.093016	2.651772	3.395257
H	4.589902	1.394446	6.003547
H	6.022945	2.335595	5.545218
H	6.482076	0.589913	3.734509
H	5.186179	-0.402397	4.432615
H	6.561711	-0.391596	6.504994
H	7.475198	-0.907609	5.210881
H	-14.953863	5.290527	-0.958641
H	-14.326246	3.919757	-1.921535
C	-14.219597	4.477971	-0.977321
H	-15.355019	3.535978	0.588188

H	-13.475511	0.859807	-2.606851
H	-12.373906	0.233416	-3.871754
N	-14.426808	3.638024	0.192710
H	-13.209576	4.897936	-0.941779
C	-12.434790	0.621232	-2.850778
N	-11.649708	1.844248	-2.746472
H	-11.874826	2.452905	-1.948516
C	-13.506684	2.739914	0.643530
H	-12.077530	-0.158545	-2.157058
C	-10.357246	1.889401	-3.168874
H	-9.914467	-0.970696	-5.605102
N	-9.931094	0.899564	-3.991241
N	-13.901847	1.946499	1.669416
N	-12.300913	2.732450	0.035587
N	-9.612893	2.935590	-2.727099
C	-8.857817	-1.160422	-5.413701
C	-8.647818	1.050340	-4.390825
H	-8.402789	-1.706724	-6.241017
N	-8.174115	0.117654	-5.289159
C	-12.936546	1.090353	2.070677
C	-11.429886	1.824888	0.560634
C	-8.339412	2.946684	-3.190478
H	-11.548735	-2.723309	-1.802708
H	-14.232909	0.324590	3.427518
N	-7.786632	2.013677	-4.019180
N	-8.775529	-1.995422	-4.211582
H	-9.391550	-1.711464	-3.451596
H	-9.958593	2.502652	-0.697234
N	-13.272867	0.256759	3.107335
N	-10.220642	1.764823	-0.036768
H	-8.803764	4.648841	-1.368648
C	-10.993108	-3.665175	-1.670749
N	-11.688680	0.968103	1.576153
H	-10.703540	-4.058350	-2.651205
H	-7.876602	5.940130	-2.185903
H	-7.158376	0.074254	-5.346181
H	-11.660513	-4.378234	-1.173187
N	-7.522742	3.972382	-2.842833
C	-7.829160	4.920267	-1.782176
H	-12.990274	-2.041299	2.035570
C	-7.550166	-2.462277	-3.806592
H	-6.535620	3.885846	-3.160846
N	-12.219446	-1.889188	2.683446
C	-12.400369	-0.778283	3.627123

C	-9.222403	0.759657	0.249639
N	-11.036321	-2.694495	0.874541
N	-6.548639	-2.440839	-4.706010
N	-9.775266	-3.478123	-0.892696
H	-9.728296	-0.167486	0.517334
H	-8.646190	0.620367	-0.672518
N	-6.823637	4.938870	-0.734061
H	-12.852911	-1.137884	4.558639
C	-10.999431	-2.126895	2.099572
C	-9.815396	-2.935854	0.346739
H	-8.895183	-3.351373	-1.421708
N	-7.585950	2.966128	0.218950
N	-7.470123	-2.922292	-2.533990
H	-5.451103	2.192431	-3.602962
H	-11.400879	-0.382313	3.830922
H	-6.120175	5.668615	-0.722619
N	-4.869734	3.017375	-3.409788
N	-8.323469	1.090877	1.340870
C	-6.692913	3.975857	0.232921
H	-4.608690	-2.749164	-6.073546
C	-5.400479	-3.011889	-4.251424
H	-4.163379	3.062718	-4.150531
C	-7.447490	2.128878	1.280647
N	-9.896212	-1.789119	2.794380
N	-4.371090	-3.052027	-5.134159
N	-8.607728	-2.667023	0.941207
C	-6.254925	-3.410195	-2.200741
H	-8.303561	0.503412	2.166983
C	-8.739064	-2.077768	2.148630
H	-4.141633	5.080232	-2.086361
C	-4.219722	2.921047	-2.086043
H	-5.018911	2.770990	-1.349990
N	-5.190952	-3.510179	-3.013901
N	-5.680057	4.152686	1.104791
N	-6.535577	2.212990	2.272800
C	-3.485987	4.235177	-1.787131
H	-6.881999	-3.602535	-0.268352
N	-6.133071	-3.884278	-0.915880
C	-3.100016	-3.720632	-4.884433
C	-5.646407	3.226046	2.112439
H	-3.516376	2.078340	-1.985313
H	-3.069124	-4.711443	-5.363249
N	-7.619735	-1.688154	2.837890
H	-2.586286	4.304910	-2.420243

H	-3.924746	4.345476	0.200904
H	-2.277992	-3.108367	-5.273122
N	-3.071780	4.306938	-0.383021
H	-7.814287	-1.232531	3.726267
H	-2.980847	-3.835577	-3.805131
C	-4.863117	-4.266803	-0.375015
C	-6.273756	-1.601918	2.305121
H	-2.509724	6.346119	-0.760258
H	-5.948962	-0.562660	2.187343
H	-6.268999	-2.092614	1.329074
N	-4.642220	3.354704	3.001912
H	-4.211303	-4.621326	-1.174171
C	-2.249475	5.491419	-0.101848
H	-3.410676	6.412276	1.467456
O	-4.234274	-3.137224	0.312576
H	-5.006107	-5.008752	0.413002
H	-3.859242	3.995506	2.748069
H	-3.748333	0.866739	1.152565
C	-3.271465	-2.422580	-0.314064
N	-3.158766	-1.180081	0.182133
C	-2.405804	5.980680	1.342918
C	-4.485610	2.472688	4.151381
H	-4.699131	1.436230	3.872632
H	-2.736084	2.375710	1.005146
C	-2.187761	-0.459988	-0.389949
H	-5.158843	2.769580	4.970178
N	-2.531528	-2.979107	-1.277323
C	-2.750281	1.300528	1.166588
O	-1.976033	0.798341	0.012263
N	-5.346334	-2.231227	3.237557
H	-1.192346	5.248818	-0.303430
C	-1.607190	-2.127635	-1.775555
N	-1.378868	-0.862513	-1.391837
N	-2.260162	4.888688	2.327884
H	-0.318188	-1.077576	-4.021833
H	-1.675991	6.795926	1.496972
N	-3.772793	-0.550913	2.967470
O	-0.860838	-2.647935	-2.758025
H	-5.499264	-3.205207	3.484466
C	-4.079061	-1.774519	3.440923
C	0.153864	-1.784912	-3.328000
H	-3.448045	2.542649	4.495443
H	-1.528076	4.217554	2.048621
H	-1.971077	5.275938	3.230605

N	-2.091331	0.950172	2.399867
H	0.833766	-2.462056	-3.846631
C	-2.472980	-0.230198	3.065266
O	-0.508222	2.566097	1.954191
H	0.676979	-1.233199	-2.545339
N	-3.249723	-2.587076	4.141797
C	-0.971583	1.743101	2.811581
N	-1.524094	-0.953266	3.677996
C	-2.005702	-2.084845	4.254524
O	-0.537515	1.577726	3.990264
H	-2.174443	-4.558794	5.271916
N	-1.080068	-2.818410	4.950065
C	-1.453555	-3.934923	5.807757
H	-0.233448	-2.298113	5.160954
H	-0.555950	-4.521606	6.033884
H	-1.917642	-3.605020	6.751027
H	1.932357	1.480784	2.079784

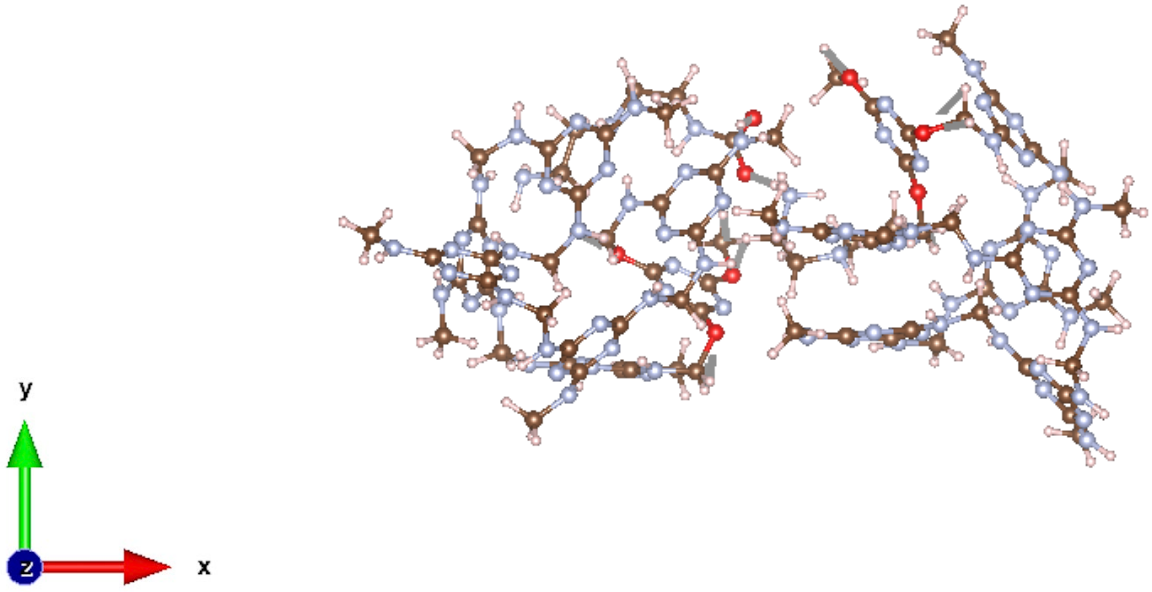

Fig. S21. DFT-calculated structure of CO_2-reacted ammonium carbamate pair within MNNs^{cya} ⊃ DETA (Structure 2). The structure was obtained by DFT calculation at the TPSS-D3(BJ)/6-31G* level.

Coordinates:

	X	Y	Z
C	1.704832	0.638247	2.868782
N	2.066754	0.520283	1.560971
C	3.393406	0.344852	1.381539
N	4.328042	0.279968	2.354260
C	3.837425	0.519887	3.580580
N	2.541492	0.668064	3.925523
N	4.743148	0.641978	4.606499
N	3.838089	0.215241	0.085479
N	0.367930	0.783002	3.126879
C	6.126413	0.963299	4.316329
C	5.133288	0.663667	-0.324653
N	6.926873	-0.252937	4.354842
C	8.225312	-0.309432	3.932911
N	8.874445	-1.448237	4.274852
C	10.128427	-1.515254	3.789751
N	10.756700	-0.579547	3.042352
C	10.004186	0.517870	2.797213
N	8.716793	0.728091	3.224648
N	10.560059	1.511982	2.070371
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