

1. Relative and Interaction Energies of Carbamic Acid Dimers

In the table below, revDSD/junTZ energies of all dimers considered in this work are reported together with the corresponding harmonic zero-point energy (ZPE) contributions, evaluated at the same level of theory. Explanation of the “revDSD/junTZ” abbreviation is provided in the manuscript.

Dimer	Ele. energy revDSD/junTZ	Harmonic ZPE	Relative energy revDSD/junTZ	Relative energy revDSD/junTZ +harmZPE
	Hartree	Hartree	kJ/mol	kJ/mol
1	-489.7269	0.10497	0.0	0
2	-489.7218	0.10539	14.5	13.9
3	-489.7181	0.10543	24.5	23.7
4	-489.7115	0.10482	40.2	39.7
5	-489.6905	0.10448	94.4	93.5

The table below collects the CP-corrected [$E_{\text{int}}(\text{CP})$] and non-CP-corrected [$E_{\text{int}}(\text{NCP})$] interaction energies at the junChS and junCBS+CV(MP2) levels. The reader is referred to the Methodology section of the manuscript for the definition of these two composite schemes.

		junChS	junCBS+CV(MP2)
		Hartree	Hartree
Dimer 1	$E_{\text{int}}(\text{CP})$	-0.0387721	-0.0387721
	$E_{\text{int}}(\text{NCP})$	-0.0391716	-0.0391716
Dimer 2	$E_{\text{int}}(\text{CP})$	-0.0307627	-0.0307627
	$E_{\text{int}}(\text{NCP})$	-0.0310608	-0.0310608
Dimer 3	$E_{\text{int}}(\text{CP})$	-0.0251922	-0.0251922
	$E_{\text{int}}(\text{NCP})$	-0.0254217	-0.0254217
Dimer 4	$E_{\text{int}}(\text{CP})$	-0.0177870	-0.0177870
	$E_{\text{int}}(\text{NCP})$	-0.0179892	-0.0179892
Dimer 5	$E_{\text{int}}(\text{CP})$	-0.0181272	-0.0181272
	$E_{\text{int}}(\text{NCP})$	-0.0183081	-0.0183081

2. Vibrational Frequencies for Carbamic Acid Dimers

In the following tables, the harmonic wavenumbers at the revDSD/junTZ level of theory are reported for all the five dimers studied in this work. As mentioned in the manuscript, for the first 4 most stable complexes (i.e. those being stable with respect to dissociation), harmonic revDSD/junTZ frequencies have been corrected for B3LYP/junDZ anharmonic contributions to obtain the hybrid anharmonic wavenumbers. Since several large amplitude motions are present, only the normal modes lying below 400 cm^{-1} have been corrected for anharmonicity. Harmonic IR Intensities are also reported as obtained from revDSD/junTZ harmonic force fields. For dimer 5, only harmonic results are reported. In the following tables, harmonic and anharmonic wavenumbers as well as anharmonic corrections are in cm^{-1} ; harmonic IR intensities are in km/mol .

Dimer 1

<i>mode</i>	<i>Harmonic Freq.</i> <i>(revDSD/junTZ)</i>	<i>Harmonic</i> <i>intensity</i>	<i>Anharmonic contribution</i> <i>B3LYP/junDZ</i>	<i>Hybrid anharm.</i> <i>Frequencies</i>
1	50.00	6.9	-	50.00
2	67.80	3.9	-	67.80
3	112.06	0.0	-	112.06
4	166.22	0.0	-	166.22
5	186.97	0.1	-	186.97
6	188.21	439.9	-	188.21
7	197.81	0.0	-	197.81
8	210.36	65.4	-	210.36
9	506.81	0.0	-39.97	466.84
10	540.54	41.0	371.90	912.44
11	555.72	0.0	133.56	689.28
12	565.79	28.5	-310.23	255.57
13	636.44	0.0	-28.58	607.86
14	652.87	27.7	0.84	653.71
15	797.53	0.0	-1.04	796.49
16	798.48	14.2	-0.10	798.38
17	954.04	0.0	-23.77	930.27
18	993.06	164.9	-1.25	991.81
19	1018.67	25.3	-27.43	991.24
20	1029.05	0.0	-105.06	923.99
21	1111.49	0.0	-28.49	1083.00
22	1115.11	20.6	-127.64	987.47
23	1387.58	0.0	-21.65	1365.93
24	1393.06	598.3	-25.30	1367.76
25	1514.43	154.8	-55.69	1458.75
26	1533.51	0.0	-107.26	1426.26
27	1614.39	0.0	-63.53	1550.86
28	1637.32	376.0	-56.61	1580.71
29	1743.38	0.0	-45.75	1697.63
30	1772.76	1302.3	-60.35	1712.41
31	3036.13	0.0	-614.04	2422.10
32	3150.91	3767.8	-571.39	2579.52
33	3639.53	110.8	-72.09	3567.44
34	3639.94	0.0	-71.32	3568.62
35	3779.10	0.2	-44.75	3734.35
36	3779.16	140.3	-43.83	3735.33

Dimer 2

<i>mode</i>	<i>Harmonic Freq. (revDSD/junTZ)</i>	<i>Harmonic intensity</i>	<i>Anharmonic contribution B3LYP/junDZ</i>	<i>Hybrid anharm. Frequencies</i>
1	49.69	8.4	-	49.69
2	60.63	3.0	-	60.63
3	99.02	0.1	-	99.02
4	145.3	1.5	-	145.3
5	168.44	3.5	-	168.44
6	184.44	40.8	-	184.44
7	259.79	218.3	-	259.79
8	399.14	193.3	48.68	447.82
9	506.86	3.7	7.72	514.58
10	525.43	61.3	-12.88	512.55
11	530.52	48.5	59.09	589.61
12	554.73	16.2	-44.17	510.56
13	615.5	34.5	-6.44	609.06
14	635.6	26.4	-7.87	627.73
15	786.88	0.34	-33.59	753.29
16	797.42	0.3	-7.57	789.85
17	804.53	48.9	-7.1	797.43
18	928.28	100.3	-51.32	876.96
19	999.1	42.2	-28.12	970.98
20	1012.5	15.4	-34.8	977.7
21	1112.77	17.5	-51.56	1061.21
22	1130.62	61.0	-27.04	1103.58
23	1252.81	133.2	-55.34	1197.47
24	1354.87	313.7	-36.19	1318.68
25	1483.22	403.7	-48.01	1435.21
26	1512.62	18.4	-56.51	1456.11
27	1617.36	139.7	-76.46	1540.9
28	1636.58	31.7	-54.04	1582.54
29	1760.31	56.5	-39.17	1721.14
30	1799.62	1121.3	-41.74	1757.88
31	3236.9	1059.0	-281.8	2955.1
32	3387.69	1233.8	-252.92	3134.77
33	3637.04	50.0	-119.26	3517.78
34	3721.59	145.2	-181.23	3540.36
35	3775.48	65.8	-117.59	3657.89
36	3813.72	100.8	-180.46	3633.26

Dimer 3

<i>mode</i>	<i>Harmonic Freq. (revDSD/junTZ)</i>	<i>Harmonic intensity</i>	<i>Anharmonic contribution B3LYP/junDZ</i>	<i>Hybrid anharm. Frequencies</i>
1	45.02	8.8	-	45.02
2	56.38	1.2	-	56.38
3	75.85	0	-	75.85
4	125.78	0	-	125.78
5	150.85	0	-	150.85
6	162.07	31.4	-	162.07
7	392.38	0	-	392.38
8	393.12	354.9	-	393.12
9	516.19	0	-5.59	510.6
10	519.61	0	-6.4	515.8
11	522.2	124.4	-4.42	515.19
12	534.22	72.9	-7.54	526.68
13	603.57	0	-5.89	597.68
14	610.65	79.2	-7.55	603.1
15	760	0	-49.07	710.93
16	786.56	4.0	-37.67	748.89
17	793.79	0	-10.76	783.03
18	805.82	118.0	-20.48	785.34
19	986.95	116.0	-27.87	959.08
20	991.03	0	-26.56	964.47
21	1121.16	120.0	-34.53	1086.63
22	1126.37	0	-36.41	1089.96
23	1248.64	318.0	-50.28	1198.36
24	1251.21	0	-50.06	1201.15
25	1464.94	485.4	-45.07	1419.87
26	1487.12	0	-47.21	1439.91
27	1644.41	0	-24.54	1619.87
28	1654.44	108.9	-22.49	1631.95
29	1777.88	0	-32.98	1744.9
30	1815.53	1092.2	-39.67	1775.86
31	3385.21	0	-234.2	3151.01
32	3427.11	1365.1	-250.53	3176.58
33	3721.25	277.7	-179.39	3541.86
34	3721.26	0	-179.34	3541.92
35	3816.81	192.5	-180.71	3636.1
36	3817.11	0	-180.09	3637.02

Dimer 4

<i>mode</i>	<i>Harmonic Freq. (revDSD/junTZ)</i>	<i>Harmonic intensity</i>	<i>Anharmonic contribution B3LYP/junDZ</i>	<i>Hybrid anharm. Frequencies</i>
1	28.57	0.06	-	28.57
2	42.68	1.4	-	42.68
3	58.66	3.5	-	58.66
4	114.73	4.0	-	114.73
5	122.55	4.5	-	122.55
6	146.78	13.6	-	146.78
7	291.05	152.2	-	291.05
8	392.41	85.7	-	392.41
9	511.37	144.3	3.24	514.61
10	511.74	7.0	-6.01	505.73
11	522.7	141.9	-30.49	492.21
12	523.8	25.9	-11.56	512.24
13	592.57	13.6	-7.24	585.33
14	604.31	52.8	-10.41	593.9
15	666.96	24.3	-45.63	621.33
16	705.64	34.3	-50.61	655.03
17	792.79	29.6	-13.26	779.53
18	799.69	1.7	-15.63	784.06
19	933.81	74.5	-25.42	908.39
20	987.91	45.9	-32.79	955.12
21	1111.74	57.9	4.94	1116.68
22	1129.65	53.7	-40.61	1089.04
23	1247.32	192.6	-43.18	1204.14
24	1252.16	102.9	-41.89	1210.27
25	1421.56	99.2	-37.65	1383.91
26	1471.7	227.6	-39.92	1431.78
27	1643.39	88.2	-34.03	1609.36
28	1668.96	80.8	-27.41	1641.55
29	1801.66	679.2	-36.79	1764.87
30	1837.75	547.4	-27.94	1809.81
31	3457.75	545.3	-117.5	3340.25
32	3567.52	365.2	-159.44	3408.08
33	3730.81	133.9	-183.45	3547.36
34	3743.75	191.2	-182.39	3561.36
35	3807.59	93.4	-186.51	3621.08
36	3814.82	100.8	-179.74	3635.08

Dimero 5

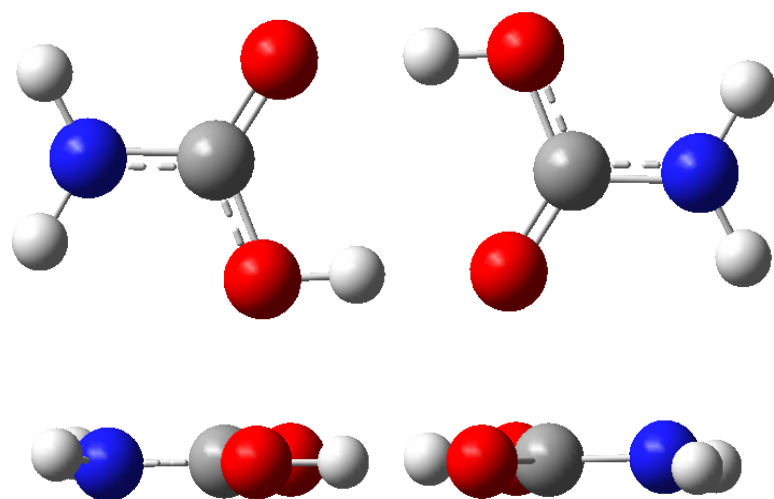
<i>mode</i>	<i>Harmonic Freq. (revDSD/junTZ)</i>	<i>Harmonic intensity</i>
1	31.50	2.9
2	44.39	7.7
3	66.28	8.1
4	89.15	5.9
5	107.17	3.2
6	125.47	12.0
7	296.91	54.3
8	422.49	227.7
9	491.44	62.9
10	520.80	146.7
11	522.62	24.5
12	537.62	59.3
13	540.53	25.8
14	614.96	3.1
15	618.23	42.6
16	634.05	64.4
17	776.70	24.3
18	789.79	6.0
19	951.34	36.4
20	957.63	50.3
21	1098.35	7.8
22	1121.85	15.3
23	1237.82	42.1
24	1283.78	63.5
25	1378.57	243.9
26	1411.04	456.4
27	1650.38	68.8
28	1670.14	68.6
29	1832.76	864.3
30	1846.12	223.0
31	3543.70	218.2
32	3605.29	60.7
33	3704.43	101.2
34	3726.87	56.8
35	3779.46	345.8
36	3832.08	76.0

3. Structures in xyz-format for Carbamic Acid Dimers

Cartesian coordinates (revDSD/junTZ) reported in the following are in Angstrom.

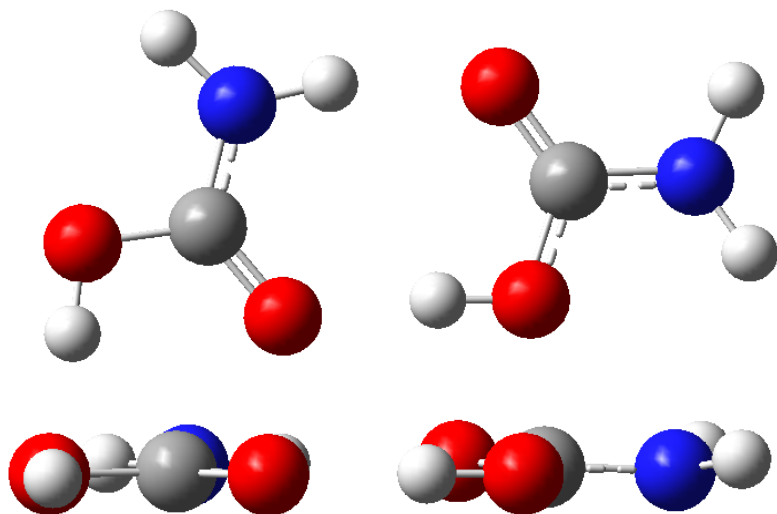
Dimer 1

C	1.886528	-0.055803	-0.002463
O	1.215611	-1.090816	-0.004168
N	3.239614	-0.075351	0.020645
H	3.700961	-0.958272	-0.093346
H	3.750737	0.779480	-0.099302
O	1.393406	1.169901	-0.003404
H	0.392435	1.124265	-0.000896
C	-1.886528	0.055803	0.002463
O	-1.215612	1.090817	0.004168
N	-3.239614	0.075350	-0.020648
H	-3.700960	0.958271	0.093355
H	-3.750735	-0.779480	0.099312
O	-1.393405	-1.169901	0.003405
N	-0.392434	-1.124265	0.000896



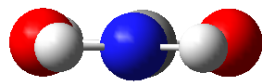
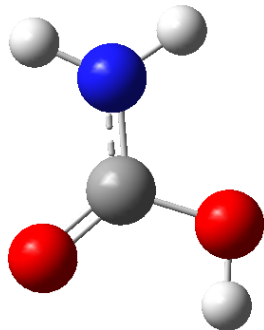
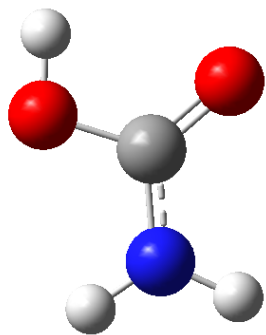
Dimer 2

C	1.965365	0.065004	0.003325
O	1.357643	1.130817	0.015659
N	3.321108	-0.009432	-0.036437
H	3.836083	0.837592	0.115893
H	3.768822	-0.894749	0.114740
O	1.403725	-1.138863	0.001332
H	0.413600	-1.051860	0.005257
C	-1.933049	-0.034124	0.000034
O	-1.255949	-1.061005	0.010522
N	-1.458799	1.213081	-0.003793
H	-0.446504	1.340753	0.001247
H	-2.095790	1.987873	-0.015348
O	-3.283775	-0.083363	-0.008444
H	-3.512768	-1.021558	-0.005481



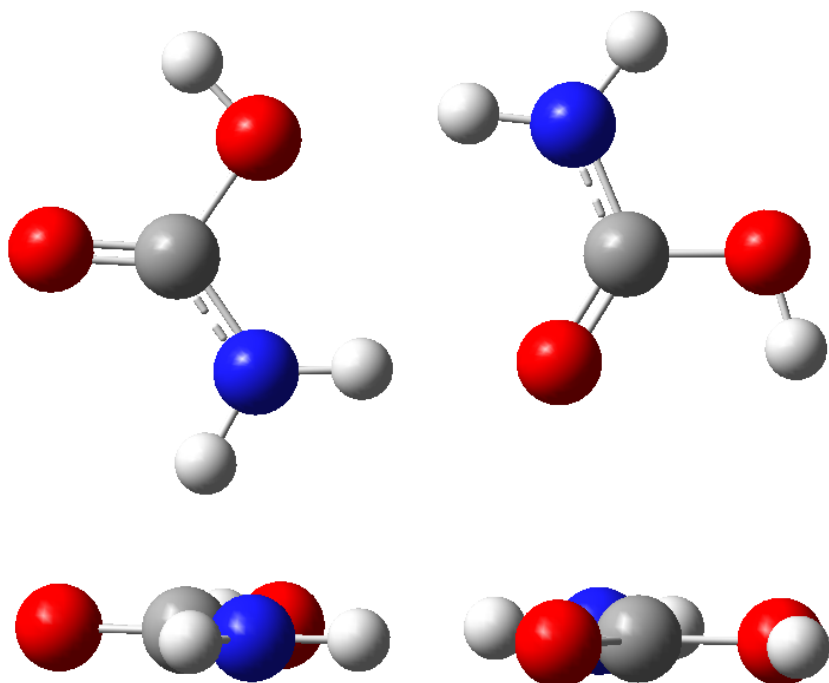
Dimer 3

C	2.013583	0.042614	0.000000
O	1.430540	1.120843	0.000000
N	1.437825	-1.165629	0.000000
H	0.421445	-1.233154	0.000000
H	2.013598	-1.987081	0.000000
O	3.367244	-0.035415	0.000000
H	3.679912	0.878015	0.000000
C	-2.013583	-0.042614	0.000000
O	-1.430540	-1.120843	0.000000
N	-1.437825	1.165629	0.000000
H	-0.421445	1.233154	0.000000
H	-2.013598	1.987081	0.000000
O	-3.367244	0.035415	0.000000
N	-3.679912	-0.878015	0.000000



Dimer 4

C	-2.069422	-0.046624	0.000000
O	-1.421261	-1.082319	0.000000
N	-1.561768	1.197225	0.000000
H	-0.559026	1.309979	0.000000
H	-2.174225	1.991346	0.000000
O	-3.422742	-0.037174	0.000000
H	-3.692293	-0.964492	0.000000
C	2.238440	-0.051321	0.000000
O	3.448860	0.020261	0.000000
N	1.486297	-1.163537	0.000000
H	1.972856	-2.040505	0.000000
H	0.471182	-1.137956	0.000000
O	1.453729	1.082153	0.000000
H	2.087841	1.810816	0.000000



Dimer 5

C	-2.105984	0.090011	-0.039789
O	-1.487922	1.094298	-0.296699
N	-3.466444	0.009007	0.049415
H	-3.958870	0.827720	-0.267830
H	-3.930326	-0.870367	-0.102570
O	-1.435872	-1.065007	0.223670
H	-2.006144	-1.743344	0.602407
C	2.243755	0.012874	0.003814
O	3.449017	-0.010280	-0.065381
N	1.505753	1.118334	0.347733
H	0.533650	1.187520	0.078755
H	2.032242	1.975021	0.355368
O	1.513478	-1.109683	-0.221405
H	0.575910	-0.955524	-0.045379

