Supplementary Material

A Cyclopropabenzenyldenethenone (Propadienone) via a New Route to Alkylidenecycloproparenes

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Contents:

P S1-S2: Experimental procedures.

P S4-S5: **Figure S1** - ¹H NMR and **Figure S2** - ¹³C NMR spectra of **12**.

P S6, Table S1: Summary computational structural data for **12** and **13**.

P S6-S15, Computational data: Cartesian coordinates, structures and energies for **12** and **13** at the B3LYP/6-31G*, B3LYP/6-311G**, and MP2/6-31G* levels, and vibrational data for **12** at the B3LYP/6-31G* level, all calculated using GAUSSIAN 98,²³ with frequencies scaled by 0.9613.²⁴

P S16, Figure S3: Full-scale experimental and calculated IR spectra (full page version of Fig. 1) of propadienone **12**.

P S17: References.

Experimental

The assignment of ¹³C and ¹H NMR resonances for new compounds was made with the aid of distortionless enhancement by polarization transfer (DEPT) and ¹H-¹H correlation spectroscopy

(COSY) experiments and confirmed by heteronuclear multiple bond connectivity (HMBC) and heteronuclear single quantum correlation (HSQC) experiments.

1-(2,2-Dimethyl-1,3-dioxane-4,6-dione-5-ylidene)-2,5-di-phenyl-1H-cyclopropabenzene **11**: To a stirred solution of Meldrum's acid¹⁴ **8** (330 mg, 2.31 mmol) under argon and in dry dichloromethane (10 mL) at ambient temperature was added pyridine (0.9 mL, 11.1 mmol). The mixture was stirred for 60 min and then solid 1,1-dichloro-2,5-diphenylcyclopropabenzene⁹ **6** (450 mg, 1.44 mmol) was added under an argon flow. The mixture, which rapidly developed a dark orange colour, was stirred at ambient temperature for 60 min and then loaded directly onto a column of silica. Elution (dichloromethane) of the orange band and recrystallization at -16°C (dichloromethane/ hexane; 1:1) of the solid obtained upon concentration gave the title exocyclic alkene **11** (125 mg, 22%) as yellow micro-crystals, mp $249\text{--}252^{\circ}\text{C}$. IR $\nu_{\text{max}}(\text{KBr})/\text{cm}^{-1}$ 3056, 2988, 1805, 1709, 1513, 1490, 1480, 1278, 1255, 1199, 1060, 927, 763, 688. UV $\lambda_{\text{max}}(\text{MeCN})/\text{nm}$ 241, 363 ($\epsilon/\text{dm}^3 \text{ mol}^{-1} \text{ cm}^{-1}$ 25 375, 39 135). ^1H NMR δ (300 MHz; CDCl_3 ; Me_4Si) 1.82 (6 H, s, $>\text{CMe}_2$), 7.48 (2 H, bd t, $J \sim 7.4 \text{ Hz}$, $\text{H}_{p/p'}$), 7.58 (4 H, bd t, $J \sim 7.6 \text{ Hz}$, $\text{H}_{m/m'}$), 8.42 (2 H, s, H_3/H_4), 8.47 (4 H, bd d, $J \sim 7.3 \text{ Hz}$, $\text{H}_{o/o'}$). ^{13}C NMR δ (75 MHz; CDCl_3 ; Me_4Si) 27.3 ($>\text{CMe}_2$), 85.9 ($\text{C}5'$), 104.3 ($\text{C}2'$), 128.5 (4 x C_o), 129.3 (4 x C_m), 129.9 ($\text{C}2/\text{C}5$), 130.3 (2 x C_p), 132.4 ($\text{C}1a/\text{C}5a$), 132.9 (2 x C_{ipso}), 136.5 ($\text{C}1$), 137.5 ($\text{C}3/\text{C}4$), 163.4 (2 x $>\text{C}=\text{O}$); μ (C_6H_6 , 21°C) 5.08 D. Found: C, 76.72; H, 4.62. Calcd. for $\text{C}_{25}\text{H}_{18}\text{O}_4 \cdot \frac{1}{2}\text{H}_2\text{O}$: C, 76.69; H, 4.89%.

Flash vacuum thermolysis of 11: The apparatus used for FVT and matrix isolation was as described previously.²² Compound **11** (10 mg, 0.03 mmol) was gently sublimed at 110°C through the FVT tube (10 cm quartz tube; 8 mm i.d) at 650°C , and the products were collected on a CsI disk cooled to 20 K in Ar matrix for 20 minutes. The system was operated at a vacuum of 2×10^{-5} mbar with a constant Ar gas flow at a flow rate of 8 mbar/min through the apparatus. The IR spectrum of the resulting Ar matrix was recorded at a resolution of $0.5\text{--}1.0 \text{ cm}^{-1}$. Wavenumbers assignable to ketene **12**: 2107 (vs), 1805 (m), 1456 (w), 1196 (w), 754 (w) cm^{-1} . Acetone: 1711 (s), 1365 (m), 1462 (w), 1228 (m), 532 (w) cm^{-1} . CO: 2138 (w), 2149 (w) cm^{-1} . CO_2 : 2340 (s), 654 (w) cm^{-1} .

The most intense bands calculated for **12** at the B3LYP/6-31G* level, using a scaling factor of 0.9613 for wavenumbers, and intensities in km/mol in parentheses, are: 2148 (2456), 1811 (360), 1460 (217), 751 (86) cm^{-1} .

When this reaction was carried out at a FVT temperature of 850°C, **12** did not survive, and a strong peak due to CO was observed instead.

Figure S1. ¹H NMR spectrum of **12** (300 MHz; CDCl₃)

Figure S2. ¹³C NMR spectrum of **12** (75 MHz; CDCl₃)

Table S1: Summary of computational structural data for **12** and **13**

	12			13		
	B3LYP/6-31G*	B3LYP/6-311G**	MP2/6-31G*	B3LYP/6-31G*	B3LYP/6-311G**	MP2/6-31G*
d (C1-C2)	1.334	1.327	1.340	1.332	1.325	1.338
d (C2-C3)	1.288	1.277	1.308	1.290	1.279	1.308
d (C3-O)	1.182	1.174	1.190	1.181	1.173	1.189
a (C1-C2-C3)	146.2	151.0	135.0	145.2	150.0	135.4
a (C2-C3-O)	171.4	173.1	169.1	171.2	173.0	169.1
torsion (phenyl anti)	2.1	13.5	24.9	-	-	-
torsion (phenyl syn)	3.9	7.5	23.3	-	-	-

2,5-Diphenylcyclopropabenzen-1-ylidenethenone 12

Symmetry: C1
Method: B3LYP/6-31G*

Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms)		
			X	Y	Z
1	6	0	0.619354	0.299412	-0.001110
2	6	0	1.377664	-0.882788	0.002474
3	6	0	0.527710	-2.008069	0.005537
4	6	0	-0.882388	-1.941542	0.005591
5	6	0	-1.620191	-0.740229	-0.000299
6	6	0	-0.751765	0.363215	-0.001345
7	1	0	0.964057	-3.002479	0.007350
8	1	0	-1.411036	-2.890102	0.014975
9	6	0	-0.017859	1.585279	-0.006355
10	6	0	-0.002234	2.919450	-0.010194
11	6	0	0.727517	3.980505	-0.012716
12	8	0	1.244186	5.044111	-0.015218
13	6	0	2.850809	-0.911490	0.002809
14	6	0	3.583061	-2.111215	0.051761
15	6	0	3.563633	0.300958	-0.045184
16	6	0	4.975470	-2.097295	0.050540
17	1	0	3.068989	-3.066393	0.094698
18	6	0	4.955518	0.313483	-0.046272
19	1	0	3.017334	1.239106	-0.082567
20	6	0	5.668975	-0.886009	0.001104
21	1	0	5.520847	-3.036321	0.089675
22	1	0	5.483454	1.262234	-0.084408
23	1	0	6.755357	-0.877567	0.000577
24	6	0	-3.088690	-0.623430	-0.000506
25	6	0	-3.934340	-1.744010	-0.082893
26	6	0	-3.677969	0.652172	0.082461
27	6	0	-5.318644	-1.593780	-0.078098
28	1	0	-3.515220	-2.742656	-0.157803
29	6	0	-5.062100	0.800097	0.087163
30	1	0	-3.042110	1.530848	0.144568
31	6	0	-5.889625	-0.322015	0.007905
32	1	0	-5.953736	-2.473042	-0.143848
33	1	0	-5.494889	1.794419	0.153114
34	1	0	-6.969980	-0.207131	0.011169

E(RB+HF-LYP) = -882.550336904

Zero-point correction=

0.258703 (Hartree/Particle)

Thermal correction to Energy=	0.276144
Thermal correction to Enthalpy=	0.277088
Thermal correction to Gibbs Free Energy=	0.208282

ModeNr.	Symmetry	Wavenumber	Abs.Int.	Rel.Int.
1	A	6,0	0,0	0
2	A	12,0	0,0	0
3	A	32,0	0,0	0
4	A	37,0	0,0	0
5	A	62,0	0,0	0
6	A	71,0	0,0	0
7	A	109,0	0,0	0
8	A	118,0	2,0	0
9	A	150,0	0,0	0
10	A	187,0	0,0	0
11	A	211,0	0,0	0
12	A	225,0	1,0	0
13	A	308,0	0,0	0
14	A	321,0	0,0	0
15	A	393,0	0,0	0
16	A	394,0	0,0	0
17	A	402,0	14,0	0
18	A	429,0	4,0	0
19	A	432,0	0,0	0
20	A	462,0	4,0	0
21	A	481,0	4,0	0
22	A	511,0	0,0	0
23	A	522,0	0,0	0
24	A	535,0	18,0	0
25	A	586,0	13,0	0
26	A	604,0	2,0	0
27	A	605,0	13,0	0
28	A	608,0	0,0	0
29	A	622,0	17,0	0
30	A	631,0	8,0	0
31	A	674,0	38,0	2
32	A	676,0	2,0	0
33	A	680,0	7,0	0
34	A	701,0	0,0	0
35	A	710,0	23,0	1
36	A	750,0	85,0	3
37	A	766,0	0,0	0
38	A	804,0	7,0	0
39	A	814,0	4,0	0
40	A	819,0	0,0	0
41	A	828,0	6,0	0
42	A	894,0	1,0	0
43	A	896,0	2,0	0
44	A	920,0	0,0	0
45	A	932,0	0,0	0
46	A	935,0	0,0	0
47	A	962,0	0,0	0
48	A	963,0	0,0	0
49	A	976,0	0,0	0
50	A	977,0	2,0	0
51	A	987,0	0,0	0
52	A	993,0	2,0	0
53	A	1025,0	2,0	0
54	A	1026,0	0,0	0
55	A	1072,0	4,0	0
56	A	1074,0	0,0	0
57	A	1109,0	12,0	0
58	A	1148,0	5,0	0
59	A	1149,0	0,0	0

60	A	1158,0	16,0	0
61	A	1167,0	23,0	1
62	A	1176,0	10,0	0
63	A	1178,0	9,0	0
64	A	1247,0	68,0	2
65	A	1267,0	16,0	0
66	A	1282,0	14,0	0
67	A	1292,0	5,0	0
68	A	1304,0	23,0	1
69	A	1322,0	0,0	0
70	A	1325,0	4,0	0
71	A	1412,0	63,0	2
72	A	1427,0	29,0	1
73	A	1442,0	11,0	0
74	A	1459,0	216,0	8
75	A	1489,0	0,0	0
76	A	1494,0	24,0	1
77	A	1517,0	4,0	0
78	A	1573,0	1,0	0
79	A	1574,0	0,0	0
80	A	1589,0	3,0	0
81	A	1597,0	1,0	0
82	A	1603,0	4,0	0
83	A	1811,0	360,0	14
84	A	2147,0	2453,0	100
85	A	3062,0	11,0	0
86	A	3063,0	3,0	0
87	A	3068,0	9,0	0
88	A	3070,0	6,0	0
89	A	3072,0	4,0	0
90	A	3078,0	1,0	0
91	A	3080,0	15,0	0
92	A	3083,0	15,0	0
93	A	3086,0	1,0	0
94	A	3089,0	21,0	1
95	A	3091,0	55,0	2
96	A	3093,0	22,0	1

Symmetry: C1
Method: B3LYP/6-311G**

Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms)		
			X	Y	Z
1	6	0	-0.627861	0.311653	0.005562
2	6	0	-1.390738	-0.864590	0.010328
3	6	0	-0.554379	-1.995879	0.026437
4	6	0	0.853911	-1.940888	0.025042
5	6	0	1.597331	-0.746622	0.014736
6	6	0	0.741594	0.363962	0.005297
7	1	0	-1.002529	-2.982331	0.052292
8	1	0	1.375989	-2.890373	0.027830
9	6	0	0.018229	1.590466	-0.008225
10	6	0	0.007159	2.917614	-0.021609
11	6	0	-0.621663	4.029168	-0.035854
12	8	0	-1.073682	5.112655	-0.048705
13	6	0	-2.863081	-0.891256	0.004351
14	6	0	-3.588901	-2.060153	-0.274679
15	6	0	-3.578474	0.285918	0.277782
16	6	0	-4.978899	-2.052577	-0.270007
17	1	0	-3.069502	-2.979716	-0.516101
18	6	0	-4.968073	0.291695	0.282949
19	1	0	-3.036770	1.199891	0.492565
20	6	0	-5.675104	-0.878117	0.010949

21	1	0	-5.520684	-2.964772	-0.492883
22	1	0	-5.499581	1.211171	0.499702
23	1	0	-6.758996	-0.874042	0.013080
24	6	0	3.066305	-0.645925	0.010311
25	6	0	3.896968	-1.765373	0.178266
26	6	0	3.670974	0.610229	-0.163945
27	6	0	5.280480	-1.633296	0.165451
28	1	0	3.466704	-2.747553	0.331649
29	6	0	5.054416	0.739958	-0.177206
30	1	0	3.048747	1.488690	-0.291484
31	6	0	5.866073	-0.381105	-0.013972
32	1	0	5.903905	-2.509990	0.299725
33	1	0	5.499302	1.718706	-0.315262
34	1	0	6.945262	-0.280218	-0.023278

E(RB+HF-LYP) = -882.759630736

Zero-point correction= 0.257325 (Hartree/Particle)
Thermal correction to Energy= 0.274778
Thermal correction to Enthalpy= 0.275722
Thermal correction to Gibbs Free Energy= 0.207526

ModeNr.	Symmetry	Wavenumber	Abs.Int.	Rel.Int.
1	A	9,0	0,0	0
2	A	20,0	0,0	0
3	A	32,0	0,0	0
4	A	33,0	0,0	0
5	A	63,0	0,0	0
6	A	68,0	0,0	0
7	A	108,0	0,0	0
8	A	114,0	2,0	7
9	A	151,0	0,0	0
10	A	185,0	1,0	3
11	A	209,0	0,0	0
12	A	226,0	1,0	3
13	A	295,0	0,0	0
14	A	330,0	0,0	0
15	A	392,0	0,0	0
16	A	394,0	0,0	0
17	A	400,0	14,0	0
18	A	423,0	7,0	0
19	A	430,0	0,0	0
20	A	465,0	3,0	0
21	A	487,0	5,0	0
22	A	509,0	0,0	0
23	A	515,0	0,0	0
24	A	539,0	18,0	0
25	A	595,0	0,0	0
26	A	604,0	2,0	7
27	A	608,0	0,0	0
28	A	609,0	26,0	0
29	A	623,0	13,0	0
30	A	629,0	12,0	0
31	A	676,0	45,0	1
32	A	677,0	18,0	0
33	A	679,0	5,0	0
34	A	703,0	1,0	3
35	A	711,0	25,0	0
36	A	748,0	96,0	3
37	A	762,0	1,0	3
38	A	799,0	9,0	0
39	A	816,0	3,0	0
40	A	818,0	0,0	0
41	A	828,0	11,0	0
42	A	898,0	0,0	0

43	A	899,0	1,0	3
44	A	931,0	0,0	0
45	A	945,0	0,0	0
46	A	946,0	0,0	0
47	A	968,0	0,0	0
48	A	969,0	0,0	0
49	A	972,0	0,0	0
50	A	976,0	1,0	3
51	A	985,0	0,0	0
52	A	992,0	6,0	0
53	A	1018,0	1,0	3
54	A	1018,0	2,0	7
55	A	1063,0	5,0	0
56	A	1065,0	0,0	0
57	A	1100,0	14,0	0
58	A	1138,0	7,0	0
59	A	1139,0	0,0	0
60	A	1149,0	29,0	1
61	A	1157,0	14,0	0
62	A	1164,0	8,0	0
63	A	1165,0	9,0	0
64	A	1232,0	68,0	2
65	A	1257,0	26,0	0
66	A	1267,0	7,0	0
67	A	1278,0	9,0	0
68	A	1286,0	26,0	0
69	A	1307,0	1,0	3
70	A	1313,0	7,0	0
71	A	1399,0	82,0	2
72	A	1413,0	22,0	0
73	A	1426,0	11,0	0
74	A	1445,0	213,0	7
75	A	1472,0	0,0	0
76	A	1480,0	35,0	1
77	A	1502,0	2,0	7
78	A	1556,0	3,0	0
79	A	1557,0	0,0	0
80	A	1575,0	1,0	3
81	A	1580,0	1,0	3
82	A	1594,0	3,0	0
83	A	1805,0	340,0	12
84	A	2157,0	2747,0	99
85	A	3042,0	7,0	0
86	A	3043,0	2,0	7
87	A	3048,0	8,0	0
88	A	3050,0	4,0	0
89	A	3052,0	6,0	0
90	A	3057,0	0,0	0
91	A	3059,0	10,0	0
92	A	3063,0	15,0	0
93	A	3066,0	2,0	7
94	A	3069,0	14,0	0
95	A	3071,0	45,0	1
96	A	3073,0	14,0	0

Symmetry: C1
Method: MP2/6-31G*

Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms)		
			X	Y	Z
1	6	0	-0.604026	0.322356	-0.003879
2	6	0	-1.336282	-0.868910	0.004907
3	6	0	-0.477928	-1.991278	0.004823

4	6	0	0.929685	-1.906608	0.001146
5	6	0	1.646715	-0.689387	0.003543
6	6	0	0.777002	0.405459	0.012379
7	1	0	-0.916954	-2.987276	0.035320
8	1	0	1.484049	-2.843110	-0.030560
9	6	0	0.022281	1.619514	0.003198
10	6	0	0.003752	2.959197	-0.003195
11	6	0	-0.934333	3.871074	-0.010528
12	8	0	-1.615057	4.846777	-0.015378
13	6	0	-2.802376	-0.921391	-0.006732
14	6	0	-3.499416	-2.038064	-0.495951
15	6	0	-3.528234	0.174424	0.490892
16	6	0	-4.893376	-2.058340	-0.480728
17	1	0	-2.954002	-2.876884	-0.922576
18	6	0	-4.921240	0.152265	0.497872
19	1	0	-2.991953	1.043972	0.866795
20	6	0	-5.608245	-0.967212	0.020111
21	1	0	-5.423694	-2.924475	-0.869451
22	1	0	-5.472041	1.006133	0.884951
23	1	0	-6.695332	-0.984403	0.026910
24	6	0	3.108329	-0.562921	0.012213
25	6	0	3.939901	-1.598350	0.468894
26	6	0	3.690886	0.627370	-0.456658
27	6	0	5.325711	-1.447104	0.449182
28	1	0	3.505013	-2.509378	0.873792
29	6	0	5.076118	0.775582	-0.468109
30	1	0	3.049688	1.436227	-0.802757
31	6	0	5.898406	-0.263745	-0.024057
32	1	0	5.960488	-2.252000	0.812174
33	1	0	5.515814	1.700837	-0.832648
34	1	0	6.979387	-0.147456	-0.034856

E(RHF) = -876.929524363
E2 = -0.2842888366D+01

Cyclopropabenzen-1-ylidenethenone 13

Symmetry: Cs
Method: B3LYP/6-31G*

Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms)		
			X	Y	Z
1	6	0	0.000000	0.697100	0.000000
2	6	0	-0.280697	2.062287	0.000000
3	6	0	-1.648940	2.345700	0.000000
4	6	0	-2.644069	1.333142	0.000000
5	6	0	-2.336898	-0.029681	0.000000
6	6	0	-0.966758	-0.287010	0.000000
7	1	0	0.470337	2.845973	0.000000
8	1	0	-1.975434	3.382457	0.000000
9	1	0	-3.686208	1.641840	0.000000
10	1	0	-3.106813	-0.794731	0.000000
11	6	0	0.406716	-0.681198	0.000000
12	6	0	1.328050	-1.643418	0.000000
13	6	0	2.591995	-1.899323	0.000000
14	8	0	3.700216	-2.307640	0.000000

E(RB+HF-LYP) = -420.428477775
Zero-point correction= 0.096865 (Hartree/Particle)
Thermal correction to Energy= 0.104654
Thermal correction to Enthalpy= 0.105598
Thermal correction to Gibbs Free Energy= 0.063335

ModeNr.	Symmetry	Wavenumber	Abs.Int.	Rel.Int.
1	A'	62,0	1,0	0
2	A"	70,0	0,0	0
3	A"	163,0	0,0	0
4	A'	253,0	3,0	0
5	A"	349,0	1,0	0
6	A'	429,0	4,0	0
7	A"	437,0	0,0	0
8	A'	523,0	16,0	0
9	A"	531,0	4,0	0
10	A'	580,0	28,0	1
11	A"	590,0	28,0	1
12	A'	677,0	5,0	0
13	A'	697,0	12,0	0
14	A"	700,0	0,0	0
15	A"	739,0	43,0	2
16	A"	863,0	0,0	0
17	A"	919,0	0,0	0
18	A"	966,0	0,0	0
19	A'	973,0	0,0	0
20	A'	991,0	0,0	0
21	A'	997,0	3,0	0
22	A'	1114,0	0,0	0
23	A'	1148,0	84,0	3
24	A'	1152,0	0,0	0
25	A'	1269,0	0,0	0
26	A'	1359,0	150,0	6
27	A'	1419,0	52,0	2
28	A'	1445,0	21,0	0
29	A'	1549,0	2,0	0
30	A'	1592,0	7,0	0
31	A'	1811,0	390,0	16
32	A'	2150,0	2435,0	100
33	A'	3067,0	1,0	0
34	A'	3079,0	6,0	0
35	A'	3094,0	20,0	1
36	A'	3097,0	15,0	0

Symmetry: Cs
Method: B3LYP/6-311G**

Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms)		
			X	Y	Z
1	6	0	0.000000	0.718438	0.000000
2	6	0	-0.298190	2.078242	0.000000
3	6	0	-1.666507	2.344822	0.000000
4	6	0	-2.647622	1.321708	0.000000
5	6	0	-2.323898	-0.033890	0.000000
6	6	0	-0.952547	-0.275005	0.000000
7	1	0	0.441513	2.869012	0.000000
8	1	0	-2.005372	3.375044	0.000000
9	1	0	-3.691063	1.617197	0.000000
10	1	0	-3.082315	-0.806626	0.000000
11	6	0	0.423894	-0.652130	0.000000
12	6	0	1.358034	-1.591830	0.000000
13	6	0	2.592159	-1.926828	0.000000
14	8	0	3.678162	-2.369473	0.000000

E(RB+HF-LYP) = -420.533412858
Zero-point correction= 0.096339 (Hartree/Particle)
Thermal correction to Energy= 0.104165
Thermal correction to Enthalpy= 0.105110

Thermal correction to Gibbs Free Energy= 0.062675

ModeNr.	Symmetry	Wavenumber	Abs.Int.	Rel.Int.
1	A'	55,0	1,0	0
2	A"	68,0	0,0	0
3	A"	161,0	0,0	0
4	A'	251,0	3,0	0
5	A"	345,0	2,0	0
6	A'	427,0	3,0	0
7	A"	433,0	0,0	0
8	A"	518,0	7,0	0
9	A'	533,0	18,0	0
10	A'	584,0	26,0	1
11	A"	609,0	26,0	1
12	A'	669,0	7,0	0
13	A'	694,0	11,0	0
14	A"	698,0	0,0	0
15	A"	735,0	58,0	2
16	A"	861,0	0,0	0
17	A"	928,0	0,0	0
18	A'	965,0	2,0	0
19	A"	973,0	0,0	0
20	A'	988,0	2,0	0
21	A'	992,0	0,0	0
22	A'	1106,0	1,0	0
23	A'	1140,0	96,0	4
24	A'	1141,0	1,0	0
25	A'	1255,0	0,0	0
26	A'	1341,0	158,0	5
27	A'	1402,0	70,0	2
28	A'	1431,0	22,0	0
29	A'	1533,0	2,0	0
30	A'	1585,0	3,0	0
31	A'	1807,0	363,0	13
32	A'	2160,0	2713,0	100
33	A'	3045,0	0,0	0
34	A'	3057,0	3,0	0
35	A'	3071,0	16,0	0
36	A'	3074,0	13,0	0

Symmetry: Cs

Method: MP2/6-31G*

Center Number	Atomic Number	Atomic Type	Coordinates (Angstroms)		
			X	Y	Z
1	6	0	0.000000	0.647218	0.000000
2	6	0	-0.256796	2.015049	0.000000
3	6	0	-1.624257	2.317349	0.000000
4	6	0	-2.633901	1.322249	0.000000
5	6	0	-2.351541	-0.049172	0.000000
6	6	0	-0.987779	-0.326299	0.000000
7	1	0	0.505536	2.789818	0.000000
8	1	0	-1.934626	3.360175	0.000000
9	1	0	-3.671895	1.648174	0.000000
10	1	0	-3.136878	-0.800416	0.000000
11	6	0	0.381567	-0.743720	0.000000
12	6	0	1.278366	-1.736277	0.000000
13	6	0	2.584482	-1.810754	0.000000
14	8	0	3.737128	-2.101452	0.000000

E(RHF) = -417.825239295

E2 = -0.1316757452D+01

Zero-point correction=

0.096683 (Hartree/Particle)

Thermal correction to Energy=

0.104661

Thermal correction to Enthalpy= 0.105605
Thermal correction to Gibbs Free Energy= 0.063042

ModeNr.	Symmetry	Wavenumber	Abs.Int.	Rel.Int.
1	A"	61,0	0,0	0
2	A'	76,0	1,0	0
3	A"	144,0	0,0	0
4	A'	258,0	3,0	0
5	A"	305,0	2,0	0
6	A"	417,0	0,0	0
7	A'	435,0	6,0	0
8	A"	472,0	6,0	0
9	A'	541,0	5,0	0
10	A"	566,0	12,0	0
11	A"	585,0	2,0	0
12	A'	589,0	32,0	2
13	A'	685,0	1,0	0
14	A"	715,0	60,0	3
15	A'	719,0	20,0	1
16	A"	841,0	0,0	0
17	A"	865,0	0,0	0
18	A"	880,0	0,0	0
19	A'	980,0	0,0	0
20	A'	997,0	2,0	0
21	A'	1002,0	6,0	0
22	A'	1127,0	2,0	0
23	A'	1144,0	67,0	3
24	A'	1177,0	1,0	0
25	A'	1281,0	0,0	0
26	A'	1395,0	10,0	0
27	A'	1450,0	64,0	3
28	A'	1456,0	29,0	1
29	A'	1570,0	2,0	0
30	A'	1586,0	15,0	0
31	A'	1852,0	377,0	17
32	A'	2149,0	2111,0	100
33	A'	3098,0	0,0	0
34	A'	3109,0	1,0	0
35	A'	3122,0	12,0	0
36	A'	3125,0	16,0	0

Figure S3. Experimental and calculated (B3LYP/6-31G*) IR spectra of propadienone **12**. Bands marked * are due to acetone. Calculated wavenumbers are scaled by 0.9613.²⁴

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