Aromatic Stabilization Energies in Excited States at the Multiconfigurational Level: Assessment in Archetypal Organic Rings

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SUPPORTING INFORMATION

Table S1. Electronic (in a.u.) and Vertical Excited state (VEE in eV) energies for benzene and fulvene at the CASSCF and NEVPT2 levels using def2–TZVP basis set. The last column indicates the experimental/computational reference values reported in the literature.

Compound	State	CASSCF	NEVPT2	VEE_{CASSCF}	VEE_{NEVPT2}	Refs. 50,51, 54–56
C_6H_6 (D_{6h})	S0	–230.8492	–231.7164	0.0	0.0	0.0
	S1	–230.6671	–231.5177	5.0	5.4	5.2
	S2	–230.5541	–231.4909	8.0	6.1	6.2
	S3	–230.5484	–231.3988	8.2	8.6	9.0
	T1	–230.7068	–231.5515	3.9	4.5	3.9
	T2	–230.6677	–231.5303	4.9	5.1	4.8
	T3	–230.6677	–231.5303	4.9	5.1	——
	T4	–230.5843	–231.5198	7.2	5.3	——
Fulvene	S0	–230.8005	–231.6613	0.0	0.0	——
	S1	–230.6466	–231.5260	4.2	3.7	3.4
	S2	–230.5751	–231.4343	6.1	6.2	5.3
	S3	–230.5301	–231.4345	7.4	6.2	——
	T1	–230.7059	–231.5650	2.6	2.6	2.4
	T2	–230.6809	–231.5390	3.3	3.3	3.1
	T3	–230.5994	–231.4487	5.5	5.8	——
	T4	–230.5639	–231.4202	6.4	6.6	——

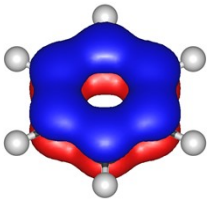
Table S2. Electronic (in a.u.) and Vertical Excited state (VEE in eV) energies for cyclobutadiene and triafulvene at the CASSCF and NEVPT2 levels using def2–TZVP basis set. The last column indicates the experimental/computational reference values reported in the literature.

Compound	State	CASSCF	NEVPT2	VEE_{CASSCF}	VEE_{NEVPT2}	Ref. 33
C_4H_4 (D_{4h})	S0	–153.7270	–154.2980	0.0	0.0	0.00
	S1	–153.6603	–154.2599	1.8	1.0	——
	S2	–153.5999	–154.2395	3.5	1.6	——
	S3	–153.3993	–154.0594	8.9	6.5	——
	T1	–153.7209	–154.2987	0.2	0.0	——
	T2	–153.5774	–154.1348	4.1	4.4	——
	T3	–153.5765	–154.1321	4.1	4.5	——
	T4	–153.3899	–154.0688	9.2	6.2	——
C_4H_4 (D_{2h})	S0	–153.7480	–154.3245	0.0	0.0	——
	S1	–153.5763	–154.2270	4.7	2.7	3.14
	S2	–153.5759	–154.1580	4.7	4.5	4.03
	S3	–153.3901	–154.0851	9.7	6.5	——
	T1	–153.6909	–154.2683	1.6	1.5	1.61
	T2	–153.5864	–154.1436	4.4	4.9	——
	T3	–153.5094	–154.0648	6.5	7.1	——
	T4	–153.3902	–154.0918	9.7	6.3	——
Triafulvene	S0	–153.7562	–154.3469	0.0	0.0	——
	S1	–153.5795	–154.1806	4.8	4.5	——
	S2	–153.4414	–154.0821	8.6	7.2	——
	S3	–153.3586	–153.9985	10.8	9.5	——
	T1	–153.6297	–154.2179	3.4	3.5	——
	T2	–153.5857	–154.1711	4.6	4.8	——
	T3	–153.4851	–154.1193	7.4	6.2	——
	T4	–153.3240	–154.0392	11.8	8.4	——

Table S3. Electronic (in a.u.) and Vertical Excited state (VEE in eV) energies for cyclooctatetraene and heptafulvene at the CASSCF and NEVPT2 levels using def2–TZVP basis set. The last column indicates the experimental/computational reference values reported in the literature.

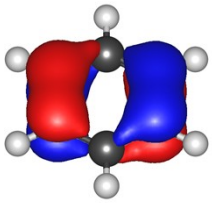
Compound	State	CASSCF	NEVPT2	VEE_{CASSCF}	VEE_{NEVPT2}	Ref. 31 and 53
C_8H_8 (D_{8h})	S0	–307.7065	–308.8397	0.0	0.0	0.0
	S1	–307.6588	–308.8190	1.3	0.6	1.4
	S2	–307.6002	–308.8128	2.9	0.7	——
	S3	–307.5233	–308.6759	5.0	4.5	——
	T1	–307.6842	–308.8349	0.6	0.1	0.7
	T2	–307.5785	–308.7124	3.5	3.5	——
	T3	–307.5784	–308.7124	3.5	3.5	——
	T4	–307.5561	–308.7027	4.1	3.7	——
C_8H_8 (D_{4h})	S0	–307.7158	–308.8527	0.0	0.0	0.0
	S1	–307.6049	–308.7982	3.0	1.5	1.8
	S2	–307.5770	–308.7557	3.8	2.6	3.0
	S3	–307.5010	–308.6569	5.8	5.3	5.8
	T1	–307.6620	–308.8127	1.5	1.1	——
	T2	–307.5738	–308.7088	3.9	3.9	——
	T3	–307.5717	–308.7087	3.9	3.9	——
	T4	–307.5180	–308.6620	5.4	5.2	——
Heptafulvene	S0	–307.7448	–308.8899	0.0	0.0	——
	S1	–307.6094	–308.7800	3.7	3.0	——
	S2	–307.5572	–308.7069	5.1	5.0	——
	S3	–307.5222	–308.6820	6.1	5.7	——
	T1	–307.6672	–308.8133	2.1	2.1	——
	T2	–307.6393	–308.7818	2.9	2.9	——
	T3	–307.5680	–308.7137	4.8	4.8	——
	T4	–307.5640	–308.7068	4.9	5.0	——

Figure S1. State Average CASSCF orbitals and electron occupations for benzene in singlet states.



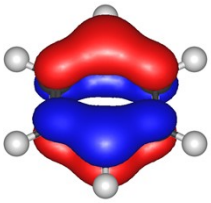
–12.9 eV

1.83 |e|



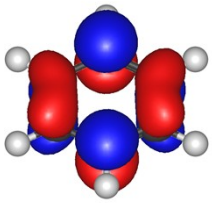
–8.1 eV

1.64 |e|



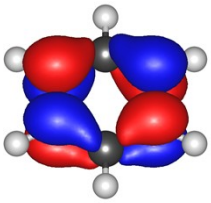
–7.6 eV

1.50 |e|



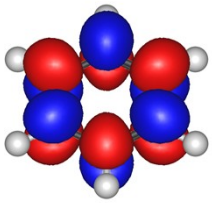
2.0 eV

0.58 |e|



2.9 eV

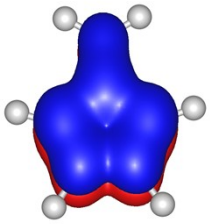
0.36 |e|



9.9 eV

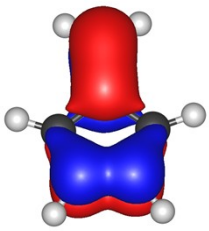
0.10 |e|

Figure S2. State Average CASSCF orbitals and electron occupations for fulvene in singlet states.



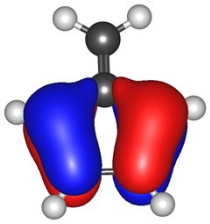
–12.8 eV

1.83 |e|



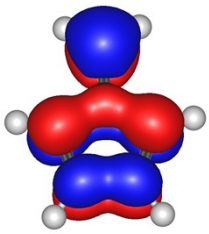
–7.8 eV

1.52 |e|



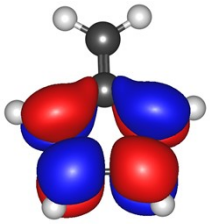
–6.6 eV

1.49 |e|



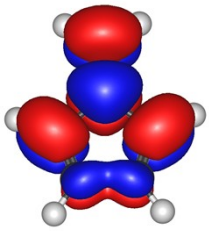
–0.9 eV

0.89 |e|



6.8 eV

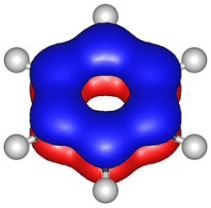
0.15 |e|



7.9 eV

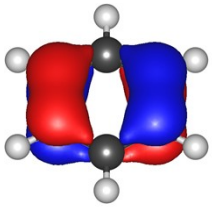
0.12 |e|

Figure S3. State Average CASSCF orbitals and electron occupations for benzene in triplet states.



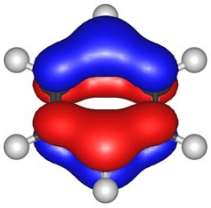
–12.9 eV

1.93 |e|



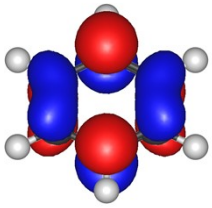
–7.6 eV

1.47 |e|



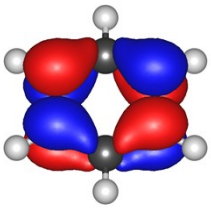
–7.6 eV

1.47 |e|



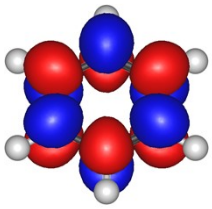
2.3 eV

0.54 |e|



2.3 eV

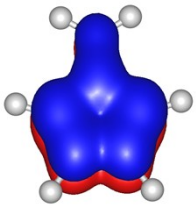
0.54 |e|



10.3 eV

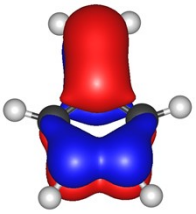
0.06 |e|

Figure S4. State Average CASSCF orbitals and electron occupations for fulvene in triplet states.



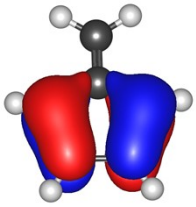
–12.7 eV

1.81 |e|



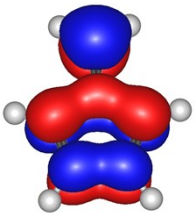
–7.7 eV

1.49 |e|



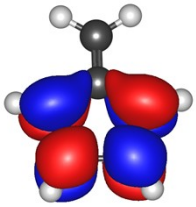
–6.2 eV

1.37 |e|



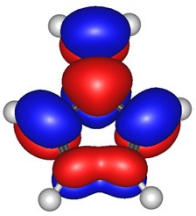
–0.7 eV

0.88 |e|



6.3 eV

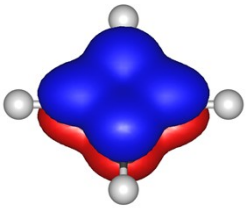
0.23 |e|



7.4 eV

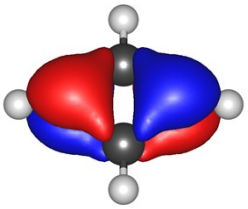
0.21 |e|

Figure S5. State Average CASSCF orbitals and electron occupations for cyclobutadiene at D_{4h} symmetry in singlet states.



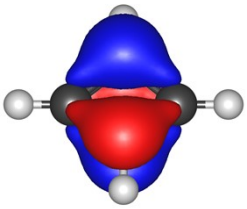
–12.6 eV

1.89 |e|



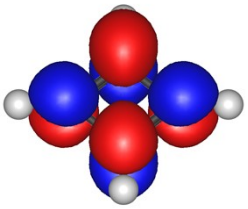
–3.0 eV

1.01 |e|



–1.8 eV

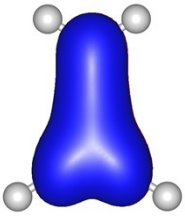
0.85 |e|



6.2 eV

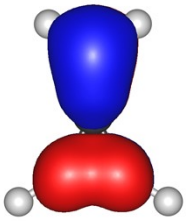
0.25 |e|

Figure S6. State Average CASSCF orbitals and electron occupations for triafulvene in singlet states.



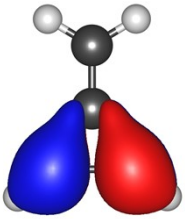
–12.7 eV

1.77 |e|



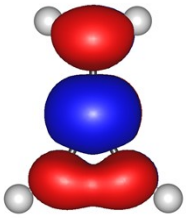
–6.0 eV

1.30 |e|



1.7 eV

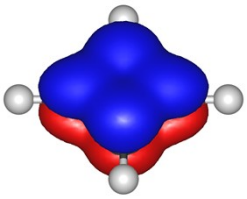
0.54 |e|



4.4 eV

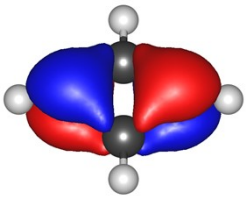
0.38 |e|

Figure S7. State Average CASSCF orbitals and electron occupations for cyclobutadiene at D_{4h} symmetry in triplet states.



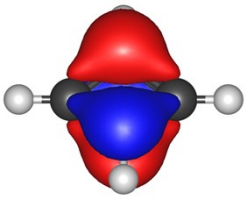
–11.5 eV

1.60 |e|



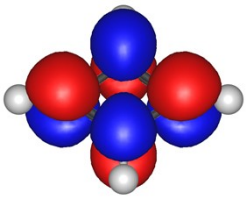
–2.8 eV

1.00 |e|



–2.0 eV

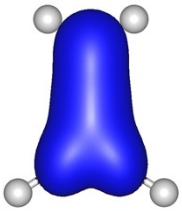
0.95 |e|



5.4 eV

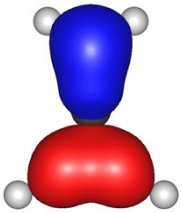
0.45 |e|

Figure S8. State Average CASSCF orbitals and electron occupations for triafulvene in triplet states.



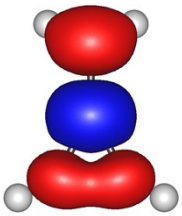
–9.9 eV

1.50 |e|



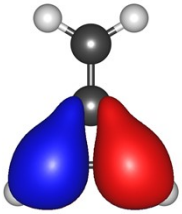
–6.5 eV

1.44 |e|



3.6 eV

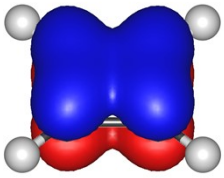
0.54 |e|



1.8 eV

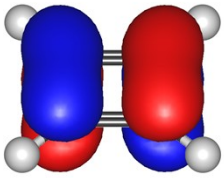
0.52 |e|

Figure S9. State Average CASSCF orbitals and electron occupations for cyclobutadiene at D_{2h} symmetry in singlet states.



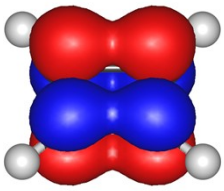
–12.3 eV

1.83 |e|



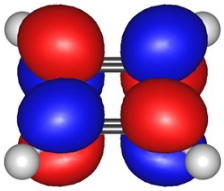
–4.4 eV

1.13 |e|



–0.75 eV

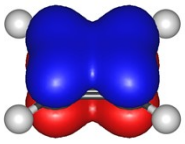
0.83 |e|



6.2 eV

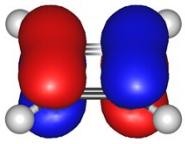
0.22 |e|

Figure S10. State Average CASSCF orbitals and electron occupations for cyclobutadiene at D_{2h} symmetry in triplet states.



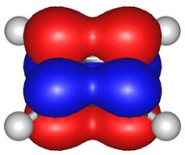
–11.2 eV

1.57 |e|



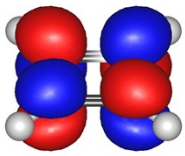
–4.3 eV

1.14 |e|



–0.8 eV

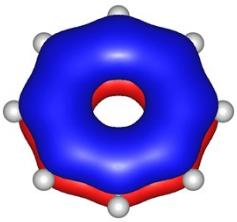
0.85 |e|



5.5 eV

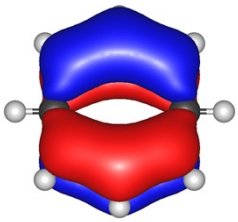
0.44 |e|

Figure S11. State Average CASSCF orbitals and electron occupations for cyclooctatetraene at D_{8h} symmetry in singlet states.



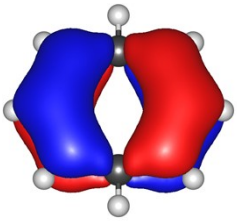
–12.8 eV

1.92 |e|



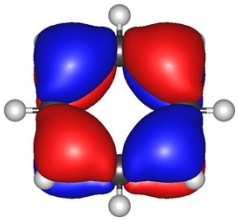
–10.3 eV

1.84 |e|



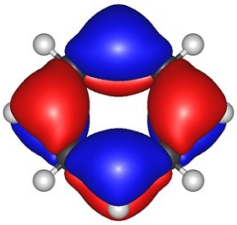
–10.3 eV

1.82 |e|



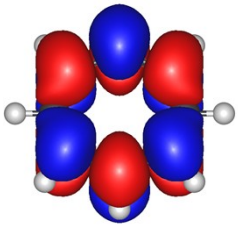
–4.3 eV

1.07 |e|



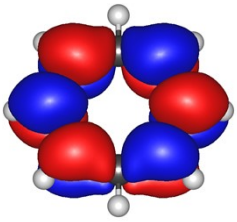
–2.3 eV

1.06 |e|



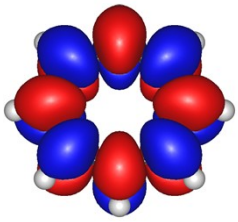
6.1 eV

0.12 |e|



6.1 eV

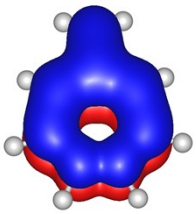
0.12 |e|



11.2 eV

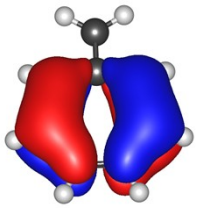
0.05 |e|

Figure S12. State Average CASSCF orbitals and electron occupations for heptafulvene in singlet states.



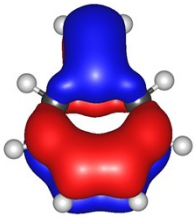
–13.0 eV

1.91 |e|



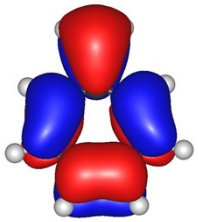
–10.2

1.81 |e|



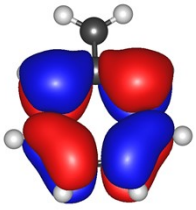
–9.6 eV

1.79 |e|



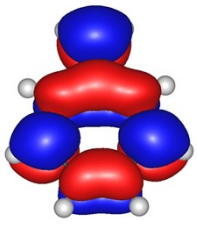
–4.6 eV

1.18 |e|



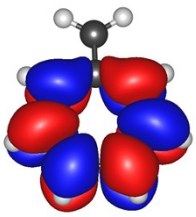
–0.5 eV

0.83 |e|



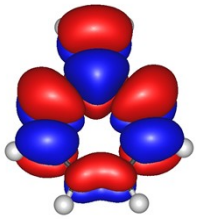
2.7 eV

0.30 |e|



8.8 eV

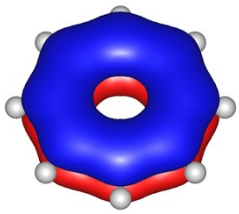
0.11 |e|



9.7 eV

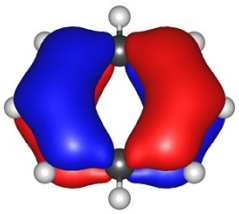
0.08 |e|

Figure S13. State Average CASSCF orbitals and electron occupations for cyclooctatetraene at D_{8h} symmetry in triplet states.



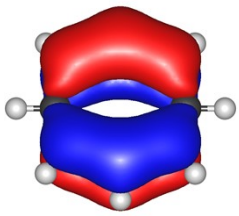
–12.5 eV

1.91 |e|



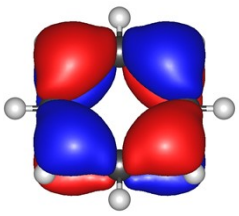
–10.1 eV

1.68 |e|



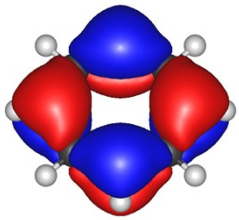
–9.6 eV

1.67 |e|



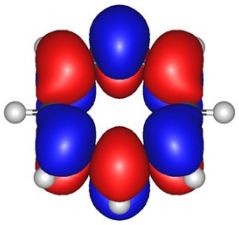
–4.6 eV

1.14 |e|



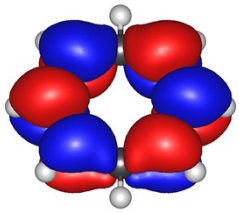
–2.0 eV

1.11 |e|



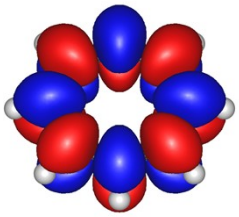
5.5 eV

0.22 |e|



5.9 eV

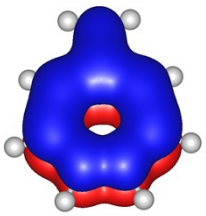
0.20 |e|



10.8 eV

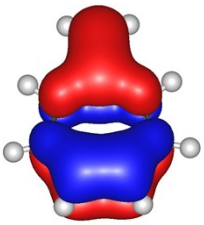
0.07 |e|

Figure S14. State Average CASSCF orbitals and electron occupations for heptafulvene in triplet states.



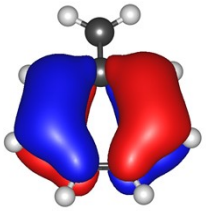
–12.9 eV

1.91 |e|



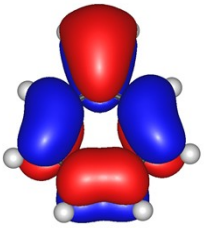
–10.0 eV

1.81 |e|



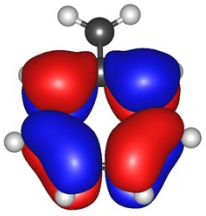
9.3 eV

1.79 |e|



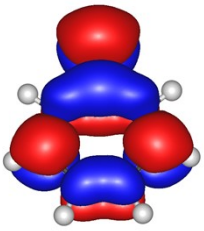
5.0 eV

1.18 |e|



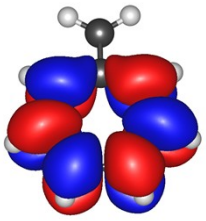
0.14 eV

0.83 |e|



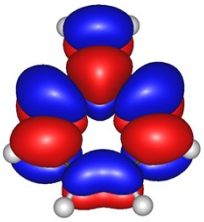
2.1 eV

0.30 |e|



8.9 eV

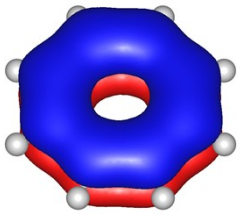
0.11 |e|



9.3 eV

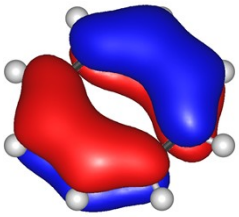
0.08 |e|

Figure S15. State Average CASSCF orbitals and electron occupations for cyclooctatetraene at D_{4h} symmetry in singlet states.



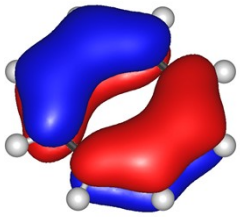
–12.8 eV

1.92 |e|



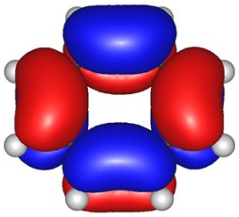
–10.3 eV

1.85 |e|



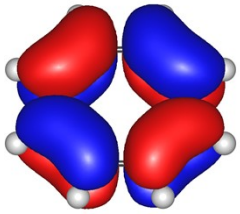
–10.3 eV

1.80 |e|



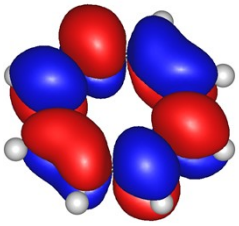
–3.4 eV

1.12 |e|



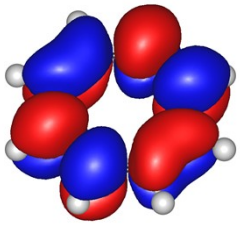
–3.2 eV

1.02 |e|



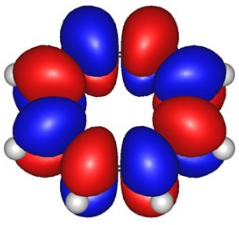
6.0 eV

0.12 |e|



6.1 eV

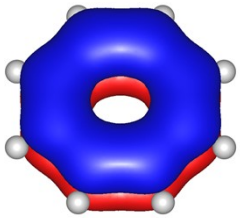
0.12 |e|



11.3 eV

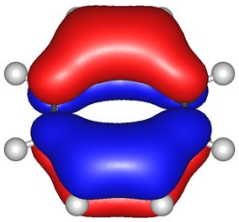
0.05 |e|

Figure S16. State Average CASSCF orbitals and electron occupations for cyclooctatetraene at D_{4h} symmetry in triplet states.



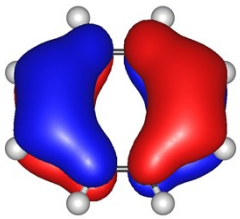
–12.5 eV

1.90 |e|



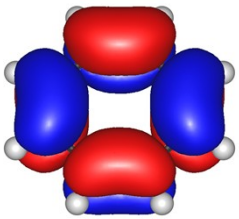
–9.8 eV

1.75 |e|



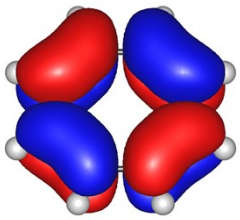
–9.8 eV

1.61 |e|



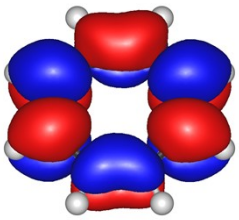
–3.4 eV

1.26 |e|



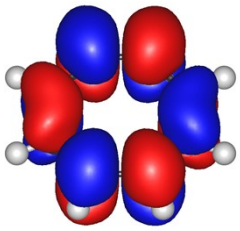
–3.2 eV

0.98 |e|



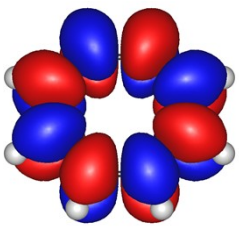
5.6 eV

0.25 |e|



5.8 eV

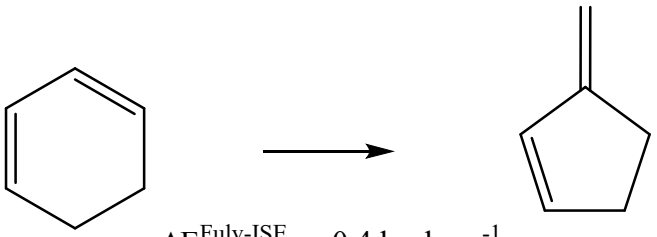
0.18 |e|



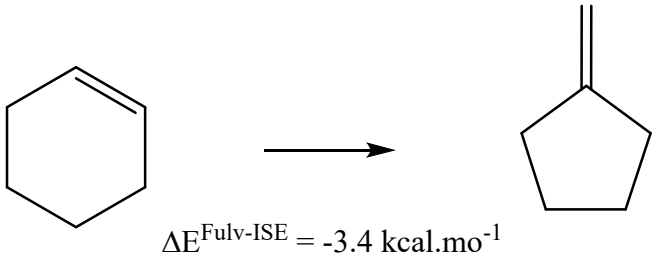
10.9 eV

0.08 |e|

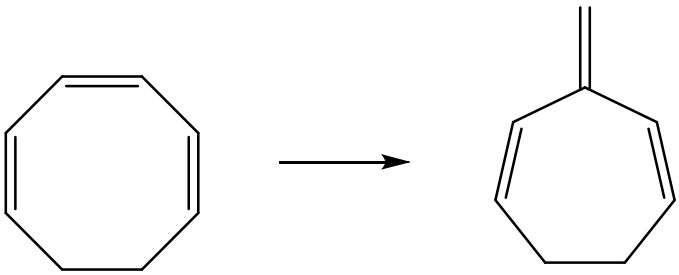
Figure S17. Isomerization stabilization reactions under the fulvenization approach for non–aromatic systems at the PBE0/def2–TZVP level.



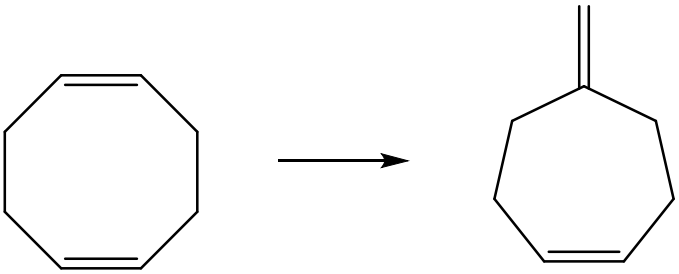
$\Delta E^{Fulv\text{-}ISE}$ = -0.4 kcal.mo^{-1}



$\Delta E^{Fulv\text{-}ISE}$ = -3.4 kcal.mo^{-1}



$\Delta E^{Fulv\text{-}ISE}$ = -4.2 kcal.mo^{-1}



$\Delta E^{Fulv\text{-}ISE}$ = -4.7 kcal.mo^{-1}

Table S4. Weight and configurations of the ground and excited states of benzene.

C_6H_6							
S_0		S_1		S_2		S_3	
0.89029	222000	0.41231	212100	0.50627	221100	0.33823	122100
0.03059	211110	0.38843	221010	0.44078	212010	0.20505	220200
0.02062	220200	0.05238	121110	0.01273	112110	0.12978	221001
0.02022	202020	0.04044	211101	0.01149	121200	0.11567	211110
0.01170	121101	0.02193	112020	0.00689	121020	0.03742	202200
0.01165	112011	0.01903	112200	0.00338	111111	0.01785	220020
		0.01747	202011	0.00326	220101	0.01525	111210
		0.01561	220011	0.00324	211011	0.01490	112011
		0.00638	012120			0.01449	210111
		0.00523	021210			0.01377	22020
		0.00422	201012			0.01302	121101
		0.00391	210102			0.01130	202020
						0.01089	120120
						0.01045	22200
						0.00837	202002
						0.00764	201201
						0.00699	220002
						0.00507	012111
						0.00294	211200
						0.00255	111012
T_1		T_2		T_3		T_4	
0.4399	221100	0.42345	212010	0.42338	221010	0.48856	212100
0.43975	212010	0.42331	221100	0.42338	212100	0.48855	221010
0.0348	122001	0.05322	112110	0.05322	121110	0.00536	201210
0.01801	111111	0.03977	211011	0.03977	211101	0.00536	210120
0.01438	201120	0.01374	121200	0.01374	112020		
0.01438	210210	0.01259	220101	0.01259	202011		
0.00634	211011	0.01050	111111	0.00359	102111		
0.00537	112110	0.00257	012210	0.00344	111021		
0.00379	120201	0.00257	021120	0.00263	120111		
0.00379	102021			0.00257	021210		
0.00317	220101			0.00257	012120		
0.00317	202101						
0.00268	121200						
0.00268	121020						

Table S5. Weight and configurations of the ground and excited states of fulvene.

Fulvene							
S_0		S_1		S_2		S_3	
0.823	222000	0.6567	221100	0.2251	122100	0.42774	212100
0.0409	212100	0.13596	211200	0.18712	220200	0.22866	202200
0.01864	202200	0.05825	221001	0.12559	212100	0.07773	122100
0.01671	121110	0.03797	121200	0.10997	221010	0.05046	212001
0.01481	211110	0.03386	211101	0.05621	112200	0.02728	211011
0.01415	220200	0.01068	202110	0.05524	211110	0.02585	222000
0.00996	202101	0.01062	112110	0.05298	220101	0.02356	221010
0.00966	112101	0.00485	201120	0.05064	202200	0.0191	22200
0.0092	220101	0.00454	210210	0.02661	220020	0.01438	111210
0.00603	220020	0.00394	121002	0.00983	121110	0.01029	202002
0.00336	221010	0.00372	211020	0.00946	201210	0.00994	122001
0.00335	202002	0.00321	211002	0.0083	112020	0.00989	121110
0.00327	121011	0.00313	111120	0.00829	210120	0.00788	112101
0.0026	112002	0.00275	210111	0.00732	121011	0.00748	220200
		0.00254	202011	0.00662	22101	0.00627	201210
				0.00531	112101	0.00598	112200
				0.00504	210201	0.0052	201111
				0.00463	111111	0.00487	211110
				0.00456	202020	0.00318	210120
				0.00385	212001	0.00286	021210
				0.00367	211011	0.00282	200220
				0.00312	222000	0.00253	111111
T_1		T_2		T_3		T_4	
0.80371	221100	0.78701	212100	0.36988	221010	0.4595	211200
0.02996	122010	0.04662	212001	0.33871	122100	0.23465	221001
0.02586	221001	0.04657	221010	0.12215	212001	0.06792	211101
0.02346	211200	0.01933	122001	0.03140	211110	0.0526	122010
0.01781	212010	0.01245	211110	0.02033	212100	0.03591	112110
0.01556	211101	0.01039	210120	0.01177	112101	0.01767	211002
0.01109	121101	0.00895	111111	0.01069	201210	0.01713	212010
0.00741	220110	0.00752	121110	0.01029	111111	0.01674	220110
0.00591	201102	0.00745	111210	0.00769	120120	0.01204	121101
0.00553	112110	0.00654	122100	0.00632	201111	0.00834	120210
0.00537	201201	0.00554	210201	0.00610	202101	0.00762	111201
0.00536	202110	0.00544	112101	0.00591	210201	0.00674	201201
0.00434	210210	0.00495	112200	0.00573	220101	0.00596	112011
0.00414	111120	0.00466	211011	0.00504	111210	0.00555	211020
0.00401	120210	0.00363	210102	0.00498	121011	0.00507	121200
0.00301	021120	0.00272	102201	0.00477	122001	0.00500	120111
0.00300	201120	0.00270	201210	0.00452	21210	0.00431	202110
0.00298	111102			0.00363	211011	0.00364	210111

Table S6. Weight and configurations of the ground and excited states of cyclobutadiene (D_{4h}).

C_4H_4 (D_{4h})							
S_0		S_1		S_2		S_3	
0.92936	2110	0.76729	2200	0.79554	2020	0.76938	2101
0.06312	1111	0.12024	2020	0.17316	2200	0.22313	1120
0.00752	0112	0.04859	0220	0.01464	2002	0.00707	1102
		0.04134	2002	0.01142	0220		
		0.01628	1201				
		0.00601	1021				
T_1		T_2		T_3		T_4	
0.92741	2110	0.57771	2101	0.49069	1210	0.51699	2011
0.06695	1111	0.35985	1120	0.44749	2011	0.47572	1210
0.00565	1120	0.03277	0121	0.03174	1012	0.00448	00211
		0.02967	1102	0.03008	0211	0.00281	1012

Table S7. Weight and configurations of the ground and excited states of cyclobutadiene (D_{2h}).

C_4H_4 (D_{2h})							
S_0		S_1		S_2		S_3	
0.89013	2200	0.997	2110	0.78332	2020	0.60148	2101
0.0467	2020			0.0627	0220	0.39583	1210
0.04139	1111			0.05816	1111	0.00257	1012
0.00904	2002			0.05482	2002		
0.00784	0220			0.03491	2200		
0.00304	0202			0.00543	0202		
T_1		T_2		T_3		T_4	
0.91678	2110	0.50882	2101	0.46236	2011	0.52807	1210
0.0546	1201	0.44796	1210	0.4407	1120	0.46871	2101
0.02269	1021	0.02172	1012	0.04993	0211		
0.00593	0112	0.0215	0121	0.047	1102		

Table S8. Weight and configurations of the ground and excited states of triafulvene.

Triafulvene							
S_0		S_1		S_2		S_3	
0.88434	2200	0.80037	2110	0.80863	2101	0.45427	1210
0.07098	2101	0.08762	1210	0.08773	2200	0.25359	2011
0.01309	1120	0.07496	2011	0.0291	1102	0.17707	2110
0.00851	2020	0.01597	0211	0.02875	2020	0.06302	211
0.00712	1102	0.01228	1111	0.0188	1201	0.02906	1111
0.00605	2002	0.00748	0112	0.00858	0121	0.02286	1012
0.00458	0202			0.00766	1120		
0.00343	0220			0.00472	1021		
				0.00341	2002		
T_1		T_2		T_3		T_4	
0.92918	2101	0.64946	2110	0.65219	1201	0.57603	1210
0.05188	1111	0.3028	1210	0.20829	2011	0.34623	2110
0.01307	0121	0.01941	1012	0.09089	1111	0.03608	1120
0.00436	2011	0.01307	1102	0.02585	1021	0.02695	1102
		0.01005	1120	0.0167	0211	0.01338	1012
		0.0052	0112	0.00499	2101		

Table S9. Weight and configurations of the ground and excited states of cyclooctatetraene (D_{8h}).

C_8H_8 (D_{8h})							
S_0		S_1		S_2		S_3	
0.80832	22211000	0.46301	22220000	0.53687	22202000	0.2652	22121000
0.04871	21211100	0.31796	22202000	0.39834	22220000	0.22959	21212000
0.04705	22111010	0.02817	20222000	0.01181	21211010	0.10815	22201010
0.02085	12211001	0.02696	22022000	0.01096	22111100	0.10614	22210100
0.00796	21111110	0.01647	22200200	0.00274	12112100	0.06932	12122000
0.00778	20211200	0.0148	22200020			0.02637	21121100
0.0075	22011020	0.01348	22111100			0.01752	22200011
0.00635	12111011	0.01236	21211010			0.01562	21201110
0.00598	11211101	0.00756	12121010			0.01	12211100
		0.00716	11221100			0.00972	20212100
		0.00679	11212010			0.00824	21112010
		0.00584	02222000			0.00823	22110110
		0.0056	21112001			0.0081	21211001
		0.00554	12112100			0.00763	21111011
		0.00488	12210110			0.0069	12111110
		0.00438	22110101			0.00612	02221010
		0.0041	21210011			0.00583	22012100
		0.00361	21201101			0.00513	21210020
		0.00337	22101011			0.0051	02212100
		0.00333	22021001			0.00406	12220010
		0.00281	12201020			0.00348	22102001
		0.00279	20221001			0.00339	22120001
						0.00336	11211020
						0.00285	12202010
						0.0028	21210200
						0.00276	11211200
						0.00272	22011101
						0.00262	12121001
						0.0026	11212001
						0.00259	20121200
						0.00253	22101002
T_1		T_2		T_3		T_4	
0.80955	22211000	0.29506	21221000	0.30838	22121000	0.26332	22112000
0.04507	22111100	0.25854	22112000	0.24409	21212000	0.24971	21221000
0.04425	21211010	0.11607	22201010	0.12378	22201100	0.13927	22210100
0.01499	12211001	0.11264	22210100	0.10434	22210010	0.10887	22201010
0.01028	21122000	0.03200	12211100	0.03347	12211010	0.04903	12211100
0.00768	22200110	0.02122	22111001	0.02404	21211001	0.03218	22111001
0.00703	21111110	0.00949	11211110	0.01014	22021100	0.01504	21112010
0.00561	11211011	0.00881	21111011	0.00972	12111110	0.01379	20221010

0.00542	12111101	0.00849	20221010	0.00904	21111101	0.01125	21201020
0.00535	22011200	0.00819	12220010	0.00836	12220100	0.0096	11211110
0.00526	20211020	0.00779	12202010	0.00777	22101200	0.00793	21210110
0.00261	21121001	0.00762	21201020	0.0071	12202100	0.00766	21111011
		0.00716	21112010	0.00644	22110110	0.00627	22110200
		0.00646	21220001	0.00633	21112100	0.00627	21121100
		0.00631	21210110	0.00506	22102001	0.00618	11222000
		0.00505	21202001	0.00504	21201110	0.00556	22012100
		0.00499	21121100	0.0048	22120001	0.00449	12112001
		0.00498	22101110	0.00465	21121010	0.00427	11221001
		0.00434	11122100	0.00439	11122010	0.00325	22101110
		0.00375	12111200	0.0036	11211020	0.00314	12111200
		0.00362	22110200	0.00335	21210200	0.00272	12210101
		0.00329	22110020	0.00320	20122001	0.00254	22011101
		0.00326	21022001	0.00300	20211011		
		0.00301	22011101	0.00299	22012010		
		0.00298	20212100	0.00296	21210020		
		0.00261	22012100	0.00264	20212010		

Table S10. Weight and configurations of the ground and excited states of cyclooctatetraene (D_{4h}).

C_8H_8 (D_{4h})							
S_0		S_1		S_2		S_3	
0.78656	22220000	0.71227	22202000	0.93596	22211000	0.32485	22121000
0.03917	22202000	0.03552	22220000	0.01145	21211100	0.18228	21212000
0.02851	21211010	0.02819	21211010	0.01046	22111010	0.11481	22210100
0.02832	22111100	0.02486	22111100	0.00339	21111110	0.09219	22201010
0.01323	12211001	0.02456	20222000	0.00334	12111101	0.06476	12122000
0.00673	21120110	0.02346	22022000	0.00312	20211020	0.01878	21112010
0.00660	11221100	0.01351	22200020	0.00308	11211011	0.01648	22200101
0.00619	21220100	0.01171	22200200	0.00307	22011200	0.01351	20221010
0.00611	22120010	0.00974	12211001			0.01121	21121100
0.00576	21121001	0.00810	12112100			0.0107	22110110
0.00527	12121010	0.00644	22102010			0.0105	21201110
0.00485	12210110	0.00613	11212010			0.00788	22021010
0.00483	20222000	0.00541	21202100			0.0069	12211100
0.00465	22022000	0.00448	02222000			0.00663	12220010
0.0042	12120101	0.00435	11221100			0.00659	02221010
0.00418	11220011	0.00431	22120010			0.00644	22111001
0.00406	22110011	0.00419	12102101			0.00627	21210200
0.00374	12221000	0.00396	21102110			0.00617	12120200
0.0037	21210101	0.00393	22012001			0.00555	21220001
0.00321	22200020	0.0039	21220100			0.005	22020101
0.0032	22200200	0.00382	21201011			0.00477	21210020
0.00311	20220020	0.00367	22101101			0.0044	02212100
0.00304	22020200	0.00356	12121010			0.00394	21111101
		0.00327	11202011			0.00388	11202110
		0.00319	12201200			0.00357	12202010
		0.00315	20212001			0.0035	21102011
		0.00305	20202020			0.00292	12111110
		0.00301	22002200			0.00281	11221001
		0.00283	21121001			0.00281	21202001
						0.00264	21210002
T_1		T_2		T_3		T_4	
0.79767	22211000	0.52249	22121000	0.50858	21221000	0.49842	22112000
0.03599	22120100	0.25100	22210100	0.22809	22210010	0.22054	22201100
0.0343	21220010	0.03064	12220100	0.04696	12220010	0.02591	22021100
0.01516	22102100	0.02049	21212000	0.03295	21220001	0.02220	22110200
0.01343	21202010	0.01963	22120001	0.02421	22112000	0.01991	12202010
0.01154	12220001	0.01684	12211010	0.01575	12211100	0.01542	21221000
0.00949	21122000	0.01212	21211001	0.01119	22111001	0.01530	21121010
0.0071	22200110	0.01064	22012100	0.00873	22201100	0.01421	12211100
0.00617	21111110	0.01004	22101200	0.00853	20212010	0.01395	21202001

0.00607	22011200	0.00575	21112010	0.00795	21201020	0.01288	12220010
0.00593	11211011	0.00565	22201010	0.00666	21112100	0.01127	22210010
0.00580	20211020	0.00430	21111011	0.00471	21111101	0.00940	12111110
0.00524	12202001	0.00410	11211110	0.00433	11211020	0.00922	21112100
0.00522	12111101	0.00394	21201110	0.00433	12111110	0.00884	22111001
0.00292	11212010	0.00393	11121011	0.00415	22101110	0.00837	21220001
		0.00364	12111200	0.00414	21210110	0.00827	21210110
		0.00360	12120110	0.00389	11220110	0.00798	21111101
		0.00355	22110110	0.00371	20211011	0.00798	12121001
		0.00346	22011101	0.00336	11212001	0.00695	22101110
		0.00341	21120101	0.00325	10222010	0.00495	12210101
		0.00333	12112001	0.00314	21120011	0.00415	11112011
		0.00314	11122010	0.00306	21121010	0.00270	20112020
		0.00300	21121100	0.00305	11121101		
		0.00275	20121020	0.00275	11122100		
		0.00264	12022100	0.00265	21021200		
				0.00265	21201200		

Table S11. Weight and configurations of the ground and excited states of heptafulvene.

Heptafulvene							
S_0		S_1		S_2		S_3	
0.74777	22220000	0.55302	22211000	0.32351	22202000	0.22452	22201100
0.03637	22210100	0.12486	22121000	0.18247	21221000	0.1814	22211000
0.02793	21221000	0.08098	22201100	0.15384	22210100	0.15037	22121000
0.02697	21211100	0.04134	22111100	0.06282	12212000	0.09776	22210010
0.01708	22202000	0.03246	22201001	0.02841	22210001	0.05668	21212000
0.01132	22111010	0.01865	12211100	0.01467	21211001	0.03689	22111100
0.01033	22200200	0.01411	12221000	0.0131	22022000	0.02058	21122000
0.00941	22110200	0.00881	22111001	0.01172	21121100	0.01935	22021100
0.00662	12210101	0.00669	21212000	0.01112	20221010	0.01884	22200110
0.00570	22112000	0.00658	21210200	0.01089	12112100	0.01882	12221000
0.00561	12211010	0.00544	20211200	0.01052	22110200	0.01279	22101200
0.00486	21211001	0.00470	22110110	0.00892	22200101	0.01194	21201110
0.00443	12120101	0.00464	21111110	0.00827	22220000	0.00926	11222000
0.0039	20222000	0.00429	21211010	0.00812	21210110	0.0077	22110110
0.00387	11221100	0.00347	22020110	0.00689	20222000	0.00632	21211010
0.00384	22110101	0.00324	22200110	0.00623	22200020	0.00586	21112100
0.00351	11221001	0.00320	21120200	0.00516	22120100	0.00544	21121010
0.00311	22020200	0.00305	21202100	0.0049	21211100	0.00542	12121001
0.00296	20221010	0.00291	20221100	0.0044	21120110	0.00514	12211001
0.00291	21210110	0.0027	21112100	0.00437	12211010	0.00503	22111001
0.00283	20220200			0.00382	21201200	0.00448	12111101
0.00268	22210001			0.00367	21201020	0.00424	20212010
0.00251	21121100			0.00366	20220200	0.00411	21210020
				0.00361	12122000	0.00325	21111110
				0.00354	11221100	0.00309	22020110
				0.0035	21220010	0.00307	21120020
				0.00295	12202100	0.00290	22011101
				0.0028	22201010	0.00270	12201101
				0.00255	21111200		
T_1		T_2		T_3		T_4	
0.69575	22211000	0.68311	22210100	0.47266	22121000	0.43551	21221000
0.05781	21221000	0.09067	22121000	0.0964	22210001	0.15455	22201100
0.02836	22201100	0.04373	21220100	0.08573	21220100	0.0783	22210010
0.0262	22120100	0.01538	12220001	0.07488	12220100	0.03181	21220010
0.01519	22120001	0.01167	22120010	0.04888	22210100	0.02881	12221000
0.01362	21211100	0.00937	22012100	0.02107	22111100	0.02666	22120100
0.01187	21220010	0.00752	22111100	0.01995	22101200	0.01698	21211100
0.00976	12220010	0.00724	21220001	0.01782	22120010	0.01605	12211100
0.00818	21201200	0.00692	12111101	0.00739	12220001	0.01478	22110200
0.00694	12211001	0.00624	21121100	0.00634	12111101	0.01349	22112000
0.00684	22112000	0.00602	21202100	0.00624	21211010	0.01085	21201200

0.006	22111010	0.00578	20212100	0.00596	12210101	0.00865	22021100
0.00545	22102100	0.00559	21211010	0.00575	12212000	0.00784	21210110
0.0052	12221000	0.00478	22101200	0.00507	12121100	0.00708	21211001
0.00486	20211200	0.00408	20211110	0.00507	21111101	0.00592	22110101
0.00475	21122000	0.00398	22110110	0.00506	21121100	0.0056	21121010
0.00419	11211101	0.00383	21111200	0.00488	21111200	0.00516	11221100
0.00408	22110200	0.0036	22011110	0.00475	21210101	0.00456	21111110
0.00365	12111110	0.0035	11211110	0.00458	22110110	0.00425	22101110
0.00353	22011200	0.00342	22200101	0.00356	21120110	0.00373	12210110
0.00342	21112100	0.00337	21201110	0.00324	22101101	0.00373	22102100
0.00323	21121010	0.00311	22101101	0.00313	21202100	0.00354	21112100
0.00315	21210110	0.0028	21212000	0.0027	11222000	0.00316	22211000
0.0031	21111110	0.00267	12210200	0.00254	21112010	0.00307	11211101
0.00283	22011101					0.00296	22111010
0.0027	21211001					0.00271	22120001

Cartesian Coordinat

C_6H_6				Fulvene			
C	–1.201542000	–0.693704000	0.000000000	C	–1.172560514	0.109181556	0.000000000
C	–1.201542000	0.693704000	0.000000000	C	0.000000000	–0.769608515	0.000000000
C	0.000000000	1.387412000	0.000000000	C	1.172560514	0.109181556	0.000000000
C	1.201542000	0.693704000	0.000000000	C	0.733320003	1.381699157	0.000000000
C	1.201542000	–0.693704000	0.000000000	C	–0.733320003	1.381699157	0.000000000
C	0.000000000	–1.387412000	0.000000000	H	–2.196935777	–0.235039972	0.000000000
H	–2.140478000	–1.235772000	0.000000000	H	1.347244199	2.272491727	0.000000000
H	–2.140478000	1.235772000	0.000000000	H	–1.347244199	2.272491727	0.000000000
H	0.000000000	2.471586000	0.000000000	H	2.196935777	–0.235039972	0.000000000
H	2.140478000	1.235772000	0.000000000	C	0.000000000	–2.105898612	0.000000000
H	2.140478000	–1.235772000	0.000000000	H	0.926057070	–2.670027652	0.000000000
H	0.000000000	–2.471586000	0.000000000	H	–0.926057070	–2.670027652	0.000000000

C_4H_4							
D_{4h}				D_{2h}			
C	0.000000000	0.000000000	1.064228000	C	0.782620000	0.000000000	0.663896000
C	0.000000000	0.954193000	0.000000000	C	–0.782620000	0.000000000	0.663896000
C	0.000000000	0.000000000	–1.064228000	C	–0.782620000	0.000000000	–0.663896000
C	0.000000000	–0.954193000	0.000000000	C	0.782620000	0.000000000	–0.663896000
H	0.000000000	0.000000000	2.135064000	H	1.546131000	0.000000000	1.429357000
H	0.000000000	2.045656000	0.000000000	H	–1.546131000	0.000000000	1.429357000
H	0.000000000	0.000000000	–2.135064000	H	–1.546131000	0.000000000	–1.429357000
H	0.000000000	–2.045656000	0.000000000	H	1.546131000	0.000000000	–1.429357000

Triafulvene			
C	0.992620076	–0.656644542	0.000000000
C	–0.280472025	0.000000000	0.000000000
C	0.992620076	0.656644542	0.000000000
H	1.565778301	–1.569807648	0.000000000
H	1.565778301	1.569807648	0.000000000
C	–1.605013057	0.000000000	0.000000000
H	–2.159661592	0.929159558	0.000000000
H	–2.159661592	–0.929159558	0.000000000

C_8H_8							
D_{8h}				D_{4h}			
C	1.302716000	1.302716000	0.000000000	C	0.668571000	1.705783000	0.000000000
C	1.302716000	–1.302716000	0.000000000	C	–1.705783000	0.668571000	0.000000000
C	0.000000000	1.805805000	0.000000000	C	1.705783000	0.668571000	0.000000000
C	–1.302716000	–1.302716000	0.000000000	C	–0.668571000	–1.705783000	0.000000000
C	1.805805000	0.000000000	0.000000000	C	–0.668571000	1.705783000	0.000000000
C	–1.805805000	0.000000000	0.000000000	C	0.668571000	–1.705783000	0.000000000
C	–1.302716000	1.302716000	0.000000000	C	1.705783000	–0.668571000	0.000000000
C	0.000000000	–1.805805000	0.000000000	C	–1.705783000	–0.668571000	0.000000000
H	2.068819000	2.068819000	0.000000000	H	1.103686000	2.701573000	0.000000000
H	2.068819000	–2.068819000	0.000000000	H	–2.701573000	1.103686000	0.000000000
H	0.000000000	2.895471000	0.000000000	H	2.701573000	1.103686000	0.000000000
H	–2.068819000	–2.068819000	0.000000000	H	–1.103686000	–2.701573000	0.000000000
H	2.895471000	0.000000000	0.000000000	H	–1.103686000	2.701573000	0.000000000
H	–2.895471000	0.000000000	0.000000000	H	1.103686000	–2.701573000	0.000000000
H	–2.068819000	2.068819000	0.000000000	H	2.701573000	–1.103686000	0.000000000
H	0.000000000	–2.895471000	0.000000000	H	–2.701573000	–1.103686000	0.000000000

Heptafulvene			
C	–0.731861242	1.557463622	0.000000000
C	0.583979360	1.275218081	0.000000000
C	1.289003913	0.000000000	0.000000000
C	0.583979360	–1.275218081	0.000000000
C	–0.731861242	–1.557463622	0.000000000
C	–1.868690330	–0.673071057	0.000000000
C	–1.868690330	0.673071057	0.000000000
H	–0.982706262	2.614396214	0.000000000
H	1.250244411	2.134409633	0.000000000
H	1.250244411	–2.134409633	0.000000000
H	–0.982706262	–2.614396214	0.000000000
H	–2.837039404	–1.164077111	0.000000000
H	–2.837039404	1.164077111	0.000000000
C	2.638074017	0.000000000	0.000000000
H	3.200959060	–0.925225123	0.000000000
H	3.200959060	0.925225123	0.000000000