

## Supplementary Material

Table 1: Summary of calculated  $\text{Pd}_n$  clusters,  $n = 4 - 309$ . Symmetry G, spin state, total energy (Hartree) and file name containing Cartesian coordinates ( $\text{\AA}$ ).

n	G	State <sup>a</sup>	Energy	File name
4	$D_{2d}$	$^3A_2$	-512.059831	pd4.1
	$T_d$	$^3T_1$	-512.059512	pd4.2
	$D_{4h}$	$^3A_{2u}$	-512.033214	pd4.3
5	$D_{3h}$	$^3A'_2$	-640.095779	pd5.1
	$D_{5h}$	$^3A''_2$	-640.033262	pd5.2
6	$D_{4h}$	$^3B_{1g}$	-768.144562	pd6.1
	$O_h$	$^3T_{2g}$	-768.143654	pd6.2
	$D_{3d}$	$^3A_{1g}$	-768.143561	pd6.3
7	$D_{5h}$	$^3A'_2$	-896.175072	pd7.1
	$C_s$	$^3A'$	-896.174055	pd7.2
	$C_1$	$^3A$	-896.173348	pd7.3
	$D_{3h}$	$^5A_1''$	-896.164777	pd7.4
	$D_{3d}$	$^5A_{1u}$	-896.157383	pd7.5
8	$D_{2d}$	$^3A_2$	-1024.223321	pd8.1
	$C_s$	$^5A'$	-1024.219442	pd8.2
	$C_s$	$^5A''$	-1024.217834	pd8.3
	$C_2$	$^5A$	-1024.217428	pd8.4
	$D_{3d}$	$^5A_{2g}$	-1024.216482	pd8.5
	$C_{2v}$	$^1A_1$	-1024.216707	pd8.6
	$C_s$	$^3A''$	-1024.215898	pd8.7
	$C_s$	$^5A''$	-1024.213969	pd8.8
	$D_{4d}$	$^1A_1$	-1024.213923	pd8.9
	$C_{2v}$	$^5B_1$	-1024.208782	pd8.10
	$T_d$	$^3A_2$	-1024.198951	pd8.11
9	$C_{2v}$	$^5A_2$	-1152.269348	pd9.1
	$C_s$	$^5A''$	-1152.267379	pd9.2
	$C_s$	$^5A_1$	-1152.266129	pd9.3
	$C_2$	$^5A$	-1152.263516	pd9.4
	$C_1$	$^5A$	-1152.262769	pd9.5

n	G	State <sup>a</sup>	Energy	File name
	$C_2$	${}^5A$	-1152.262575	pd9.6
	$C_{2v}$	${}^5B_2$	-1152.260313	pd9.7
	$D_{3h}$	${}^1A_1$	-1152.259949	pd9.8
	$C_{2v}$	${}^5A_1$	-1152.257014	pd9.9
10	$D_{2h}$	${}^5A_{2g}$	-1280.320755	pd10.1
	$C_{3v}$	${}^7A_2$	-1280.320010	pd10.2
	$C_{2v}$	${}^3B_2$	-1280.315265	pd10.3
	$C_s$	${}^7A'$	-1280.313569	pd10.4
	$C_{3v}$	${}^5A_1$	-1280.302955	pd10.5
	$T_d$	${}^9A_2$	-1280.298719	pd10.6
	$C_{4v}$	${}^3B_2$	-1280.294800	pd10.7
	$T_d$	${}^3A_2$	-1280.294420	pd10.8
	$D_{5d}$	${}^5A_{1u}$	-1280.268205	pd10.9
11	$C_s$	${}^7A''$	-1408.372796	pd11.1
	$D_{3h}$	${}^7A'_2$	-1408.371953	pd11.2
	$C_1$	${}^5A$	-1408.367953	pd11.3
	$C_s$	${}^9A'$	-1408.366116	pd11.4
	$C_s$	${}^7A''$	-1408.365439	pd11.5
	$C_{2v}$	${}^7A_2$	-1408.364602	pd11.6
	$C_{2v}$	${}^7B_2$	-1408.364443	pd11.7
	$C_s$	${}^7A'$	-1408.363361	pd11.8
	$C_1$	${}^1A$	-1408.360111	pd11.9
	$C_2$	${}^3B$	-1408.356524	pd11.10
	$D_{5d}$	${}^5E_{2u}$	-1408.350580	pd11.11
	$C_s$	${}^5A'$	-1408.346146	pd11.12
	$D_{3h}$	${}^1A_1$	-1408.341223	pd11.13
	$D_{2h}$	${}^5A_{2u}$	-1408.335600	pd11.14
	$T_d$	${}^3T_1$	-1408.296506	pd11.15
12	$D_{2d}$	${}^7B_2$	-1536.429332	pd12.1
	$C_1$	${}^9A$	-1536.422695	pd12.2
	$C_s$	${}^9A''$	-1536.413596	pd12.3
	$C_s$	${}^9A''$	-1536.413595	pd12.4
	$C_{5v}$	${}^5A_1$	-1536.408222	pd12.5
	$D_{3d}$	${}^3A_{2g}$	-1536.407579	pd12.6
	$O_h$	${}^7A_{1u}$	-1536.353518	pd12.7

n	G	State <sup>a</sup>	Energy	File name
13	$D_{3d}$	${}^9A_{2u}$	-1664.470686	pd13.1
	$I_h$	FON	-1664.465815	pd13.2
	$C_{2v}$	${}^9A_2$	-1664.465153	pd13.3
	$D_{4h}$	${}^5B_{2g}$	-1664.462316	pd13.4
	$D_{5h}$	${}^9E''_1$	-1664.461564	pd13.5
	$D_{3h}$	${}^7A'_2$	-1664.458542	pd13.6
	$C_{2v}$	${}^3B_1$	-1664.455237	pd13.7
	$O_h$	${}^7A_{1u}$	-1664.447901	pd13.8
15	$C_{2v}$	${}^9B_2$	-1920.586779	pd15.1
	$O_h$	${}^9A_{1g}$	-1920.581269	pd15.2
16	$C_s$	${}^9A'$	-2048.646335	pd16.1
	$C_3$	${}^3A$	-2048.634468	pd16.2
19	$O_h$	${}^9E_g$	-2432.846025	pd19.1
	$D_{5h}$	${}^9A'_1$	-2432.806386	pd19.2
23	$D_{5h}$	${}^{15}A''_1$	-2945.065793	pd23.1
	$D_{3h}$	${}^9A''_1$	-2945.034895	pd23.2
	$D_{3h}$	${}^9A'_1$	-2945.031923	pd23.3
25	$C_{2v}$	${}^{11}B_1$	-3201.142426	pd25.1
26	$D_{3h}$	${}^9A'_1$	-3329.216945	pd26.1
	$T_d$	${}^3A_2$	-3329.186285	pd26.2
27	$C_{2v}$	${}^{11}B_2$	-3457.251177	pd27.1
29	$D_{3h}$	${}^{11}A'$	-3713.355336	pd29.1
35	$C_3$	${}^{15}A$	-4481.719466	pd35.1
38	$O_h$	${}^9A_{1g}$	-4866.003383	pd38.1
39	$D_{5h}$	${}^{11}A'_2$	-4994.055066	pd39.1
	$C_2$	${}^{19}A$	-4994.037734	pd39.2
44	$O_h$	${}^1A_1$	-5634.381883	pd44.1
54	$D_{5h}$	${}^{11}E'_1$	-6914.980362	pd54.1
55	$C_{2h}$	${}^{15}B_g$	-7043.088355	pd55.1
	$I_h$	FON	-7043.079388	pd55.2
	$O_h$	${}^{13}A_{2g}$	-7043.077470	pd55.3
	$D_{5h}$	${}^{17}E''_2$	-7043.074104	pd55.4
57	$D_{3h}$	${}^{17}A''_2$	-7299.188908	pd57.1
59	$D_3$	${}^{27}E$	-7555.244565	pd59.1
65	$O_h$	${}^{35}A_{2u}$	-8323.755635	pd65.1

n	G	State <sup>a</sup>	Energy	File name
75	$D_{5h}$	$^{33}E_2''$	-9604.483690	pd75.1
85	$O_h$	$^{23}A_{2u}$	-10885.164475	pd85.1
	$D_{5h}$	$^{25}A_2''$	-10885.162744	pd85.2
105	$D_{5h}$	FON	-13446.464607	pd105.1
115	$I_h$	FON	-14727.007233	pd115.1
116	$O_h$	FON	-14855.337239	pd116.1
140	$O_h$	$^7A_{2g}$	-17929.106099	pd140.1
145	$I_h$	FON	-18569.054117	pd145.1
146	$O_h$	$^1A_1$	-18697.568379	pd146.1
	$D_{5h}$	FON	-18697.519561	pd146.2
147	$I_h$	FON	-18825.513069	pd147.1
	$O_h$	FON	-18825.511688	pd147.2
	$D_{5h}$	FON	-18825.447434	pd147.3
231	$O_h$	FON	-29583.701444	pd231.1
309	$O_h$	FON	-39573.446446	pd309.1
	$I_h$	FON	-39573.430754	pd309.2

<sup>a</sup> FON = fractional occupation numbers.