ARTICLE

Supporting Information for

"Model calculations for the prediction of the diradical character of physisorbed molecules: p-Benzyne/MgO and p-Benzyne/SrO"

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Table S1. Spin contamination errors (SCEs) in the total energies of the calculated models.

Model	*SCE* [eV]
Model A; Fig. 2(a)	0.198
Model B; Fig. 2(b)	0.188
Model C; Fig. 2(c)	0.181
Model D; Fig. 2(d)	0.175
Model E; Fig. 2(e)	0.179
Model A'; Fig. 10(a,b)	0.199
Model B'; Fig. 10(c,d)	0.192
Model C'; Fig. 10(e,f)	0.184
Model D'; Fig. 10(g,h)	0.072
Model E'; Fig. 10(i,j)	0.147
Benzyne; Fig. 4(a)	0.209

Table S2. E_{dis} values with ($E_{dis,\ AP}$) and without ($E_{dis,\ DFT}$) spin contamination error correction

Model	$E_{dis,\ AP}$ [meV]	$E_{dis,\ DFT}$ [meV]
Model A; Fig. 2(a)	2	1
Model B; Fig. 2(b)	5	9
Model C; Fig. 2(c)	–3	4
Model D; Fig. 2(d)	–1	4
Model E; Fig. 2(e)	–1	12

Table S3. Local magnetic moment (μ_B [e$^-$]) of atoms in benzyne in models without surfaces

Atoms[a]	Model A	Model B	Model C	Model D	Model E
C1	0.037	0.035	0.036	0.036	0.035
C2	–0.035	–0.035	–0.036	–0.036	–0.035
C3	0.442	0.440	0.440	0.440	0.439
C4	–0.037	–0.035	–0.036	–0.036	–0.035
C5	0.036	0.035	0.036	0.036	0.035
C6	–0.442	–0.440	–0.440	–0.440	–0.439
H1	–0.008	–0.008	–0.008	–0.008	–0.008
H2	0.008	0.008	0.008	0.008	0.008
H3	0.008	0.008	0.008	0.008	0.008
H4	–0.008	–0.008	–0.008	–0.008	–0.008

[a] Atom labelling is given in Fig. 4(a), and C3 and C6 atoms are main spin sites.

Table S4. Local magnetic moments (μ_B [e$^-$]) of atoms in benzyne models with surfaces

Atoms[a]	Model A	Model B	Model C	Model D	Model E
C1	0.035	0.035	0.035	0.032	0.032
C2	–0.036	–0.027	–0.033	–0.032	–0.031
C3	0.437	0.425	0.416	0.387	0.406
C4	–0.035	–0.027	–0.034	–0.031	–0.032
C5	0.036	0.035	0.033	0.032	0.031
C6	–0.438	–0.405	–0.417	–0.415	–0.408
H1	–0.008	–0.005	–0.007	–0.007	–0.007
H2	0.007	0.009	0.007	0.007	0.007
H3	0.008	0.009	0.007	0.007	0.007
H4	–0.007	–0.005	–0.007	–0.007	–0.007

[a] Atom labelling is given in Fig. 4(a), and C3 and C6 atoms are main spin sites

Table S5. Local magnetic moments (μ_B [e⁻]) of atoms in models of MgO or SrO with benzyne adsorptions

Atoms[a]	Model A	Model B	Model C	Model D	Model E
A1	0.000	0.000	0.000	0.002	0.000
A2	0.000	0.000	0.000	0.000	0.000
A3	0.000	0.000	0.000	0.000	0.000
A4	0.000	0.000	0.000	0.000	0.000
A5	0.000	0.000	0.000	0.000	0.000
A6	0.000	0.000	0.000	0.000	0.000
A7	0.000	0.000	0.000	0.000	0.000
A8	0.000	0.000	0.000	0.000	0.000
A9	0.000	0.000	0.000	0.000	0.000
A10	0.000	0.000	0.000	0.000	0.000
A11	0.000	0.000	0.001	0.000	0.000
A12	0.000	0.000	0.000	0.002	0.002
A13	0.000	0.000	0.000	0.000	0.000
A14	0.000	0.000	0.000	0.000	0.000
A15	0.000	0.000	-0.001	0.000	-0.002
A16	0.000	0.000	0.000	0.000	0.000
A17	0.000	0.000	0.000	0.009	0.000
A18	0.000	0.000	0.000	0.000	0.000
A19	0.000	0.000	0.000	0.000	0.000
A20	0.000	0.000	0.000	0.000	0.000
A21	0.000	0.000	0.000	0.000	0.000
A22	0.000	0.000	0.000	0.000	0.000
A23	0.000	0.000	0.000	0.000	0.000
A24	0.000	-0.003	0.000	0.000	0.000
A25	0.000	0.000	0.000	-0.003	0.000
A26	0.000	0.001	0.000	0.001	0.000
A27	0.000	0.001	0.014	0.001	0.000
A28	0.000	0.000	0.015	0.035	0.061
A29	0.000	0.001	0.000	0.001	-0.001
A30	0.000	0.000	-0.012	-0.003	0.000
A31	0.000	0.000	-0.015	0.000	-0.054
A32	0.000	0.001	0.000	0.001	0.000
O1	0.000	0.000	0.000	0.000	0.000
O2	0.000	0.000	0.000	0.000	0.000
O3	0.000	0.000	0.000	0.000	0.000
O4	0.000	0.000	0.000	0.000	0.000
O5	0.000	0.000	0.000	0.000	0.000
O6	0.000	0.000	0.000	0.000	0.000
O7	0.000	0.000	0.000	0.000	0.000
O8	0.000	0.000	0.000	0.000	0.000
O9	0.000	0.000	0.000	0.000	0.000
O10	0.000	0.000	0.000	0.000	0.000
O11	0.000	0.000	0.000	0.000	0.000
O12	0.000	0.000	0.000	0.000	0.000
O13	0.000	0.000	0.000	0.000	0.000
O14	0.000	0.000	0.000	0.000	0.000
O15	0.000	0.000	0.000	0.000	0.000
O16	0.000	0.000	0.000	0.000	0.000
O17	0.000	0.000	0.000	0.000	0.000
O18	0.000	-0.002	0.000	0.000	0.000
O19	0.000	-0.002	0.000	0.000	0.000
O20	0.000	0.000	0.000	0.001	0.002
O21	0.001	-0.002	0.000	0.000	0.000
O22	0.000	0.000	0.000	0.000	0.000
O23	0.000	0.000	0.000	0.000	-0.002
O24	0.000	-0.002	0.000	0.000	0.000

O25	0.014	0.000	0.001	0.002	0.000
O26	0.000	0.001	0.000	0.000	0.001
O27	-0.010	0.000	0.000	0.000	0.000
O28	0.000	0.000	-0.001	0.000	-0.001
O29	0.000	0.001	0.000	0.000	0.000
O30	0.000	0.000	0.000	0.000	0.000
O31	0.000	0.001	0.000	0.000	0.000
O32	0.000	-0.097	0.000	0.000	0.000

[a] A1-A32 are Mg or Sr atoms, and O1-O64 are O atoms.

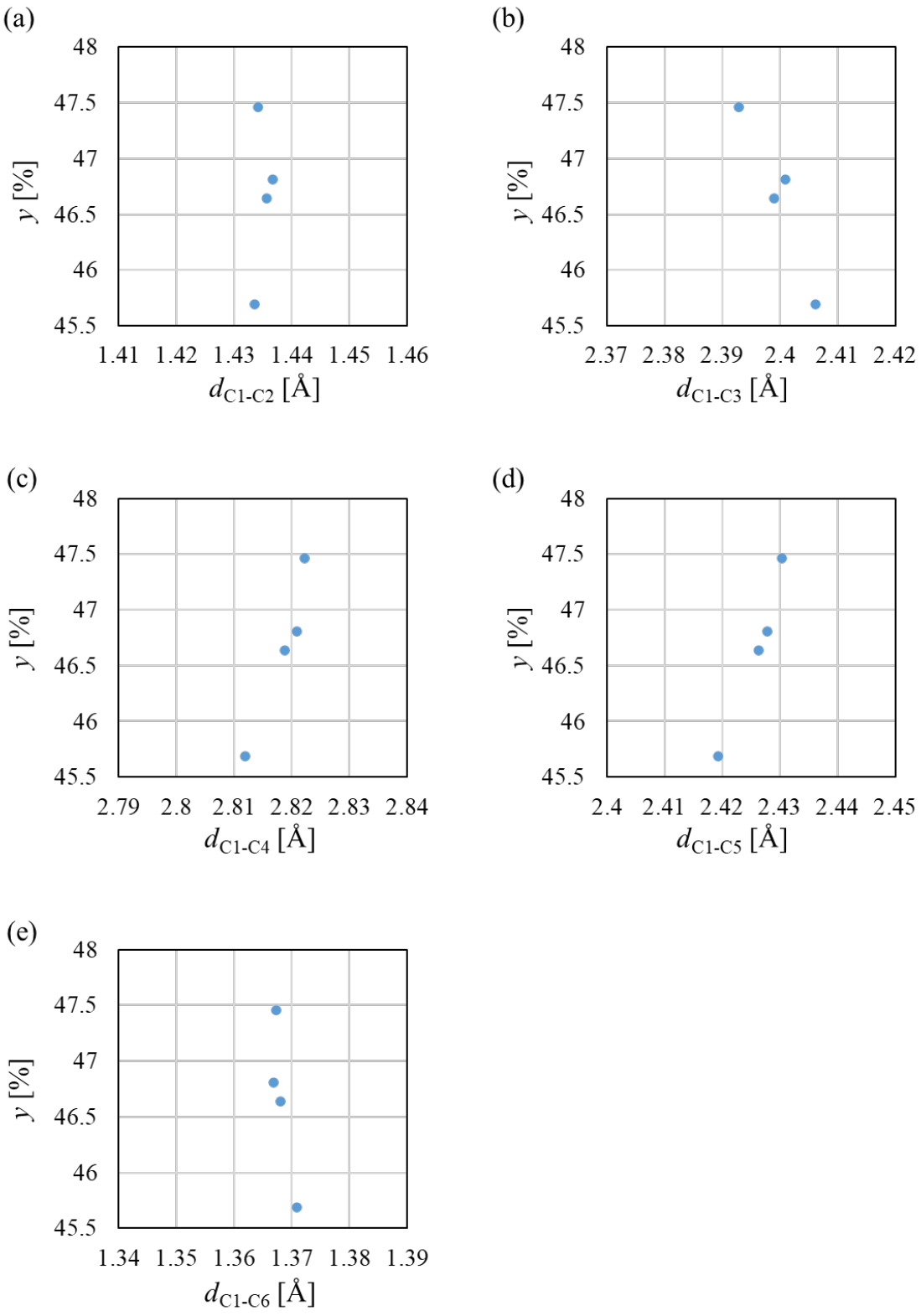

Figure S1. The plotted diradical character (y) against distances (a) $d_{C1\text{-}C2}$, (b) $d_{C1\text{-}C3}$, (c) $d_{C1\text{-}C4}$, (d) $d_{C1\text{-}C5}$, and (e) $d_{C1\text{-}C6}$, where $d_{A\text{-}B}$ refers to the distance between atoms A and B, and indices for C atoms are shown in the main text (Fig. 4(a)).

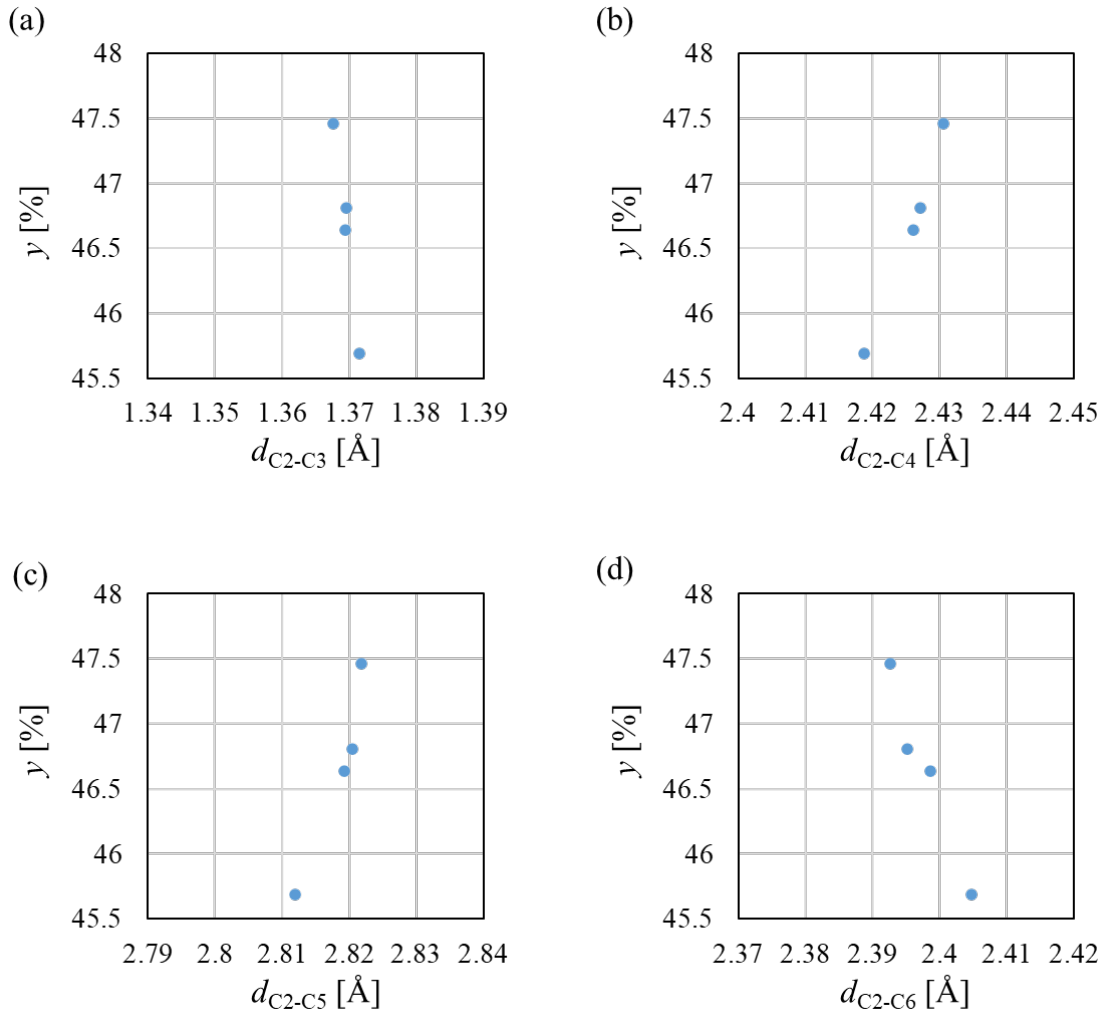

Figure S2. The plotted diradical character (*y*) against distances (a) $d_{C2\text{-}C3}$, (b) $d_{C2\text{-}C4}$, (c) $d_{C2\text{-}C5}$, and (d) $d_{C2\text{-}C6}$, where $d_{A\text{-}B}$ refers to the distance between atoms A and B, and indices for C atoms are shown in the main text (Fig. 4(a)).

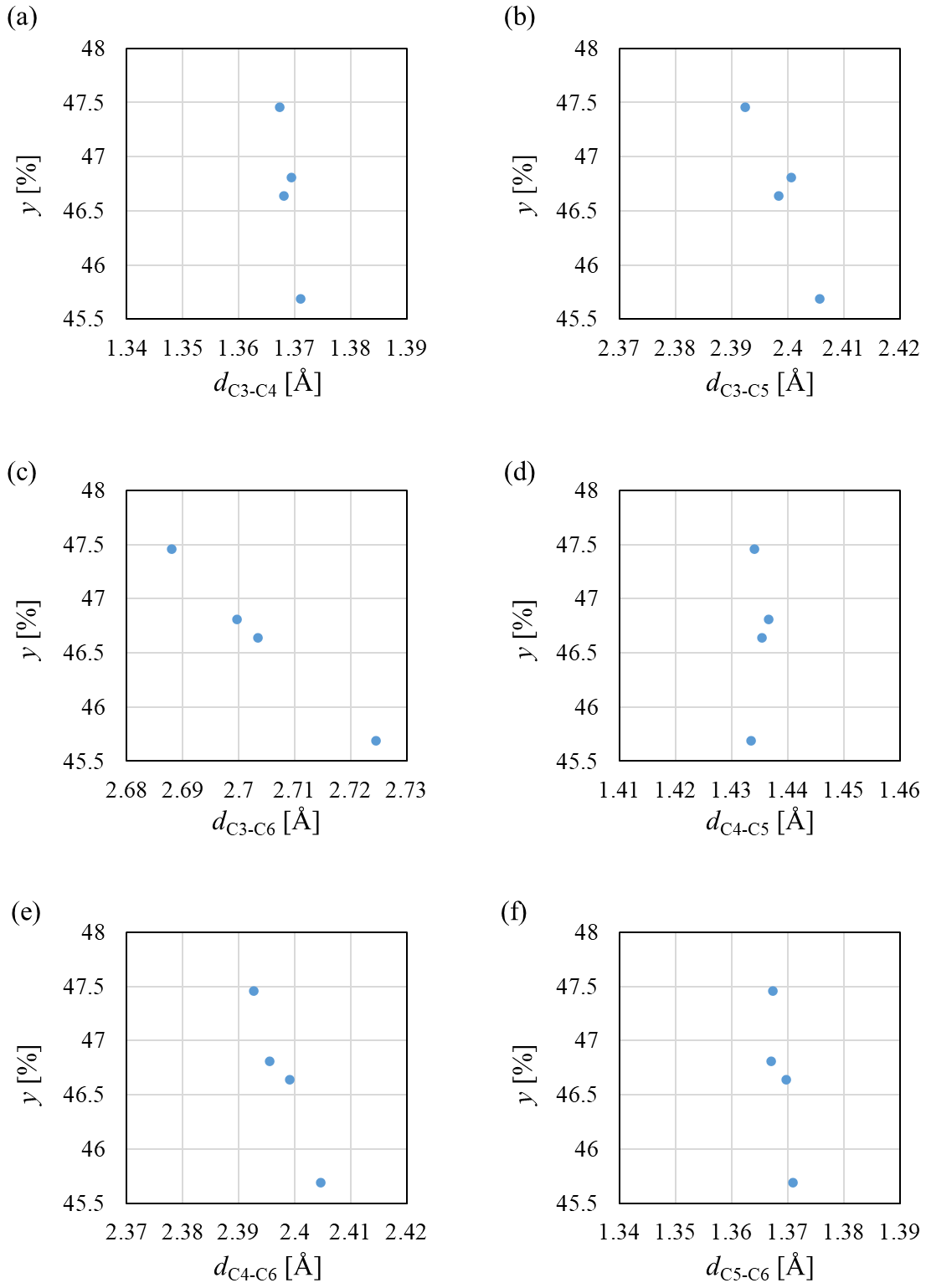

Figure S3. The plotted diradical character (*y*) against distances (a) $d_{C3\text{-}C4}$, (b) $d_{C3\text{-}C5}$, (c) $d_{C3\text{-}C6}$, (d) $d_{C4\text{-}C5}$, (e) $d_{C4\text{-}C6}$, and (f) $d_{C5\text{-}C6}$, where $d_{A\text{-}B}$ refers to the distance between atoms A and B atoms, and indices for C atoms are shown in the main text (Fig. 4(a)).

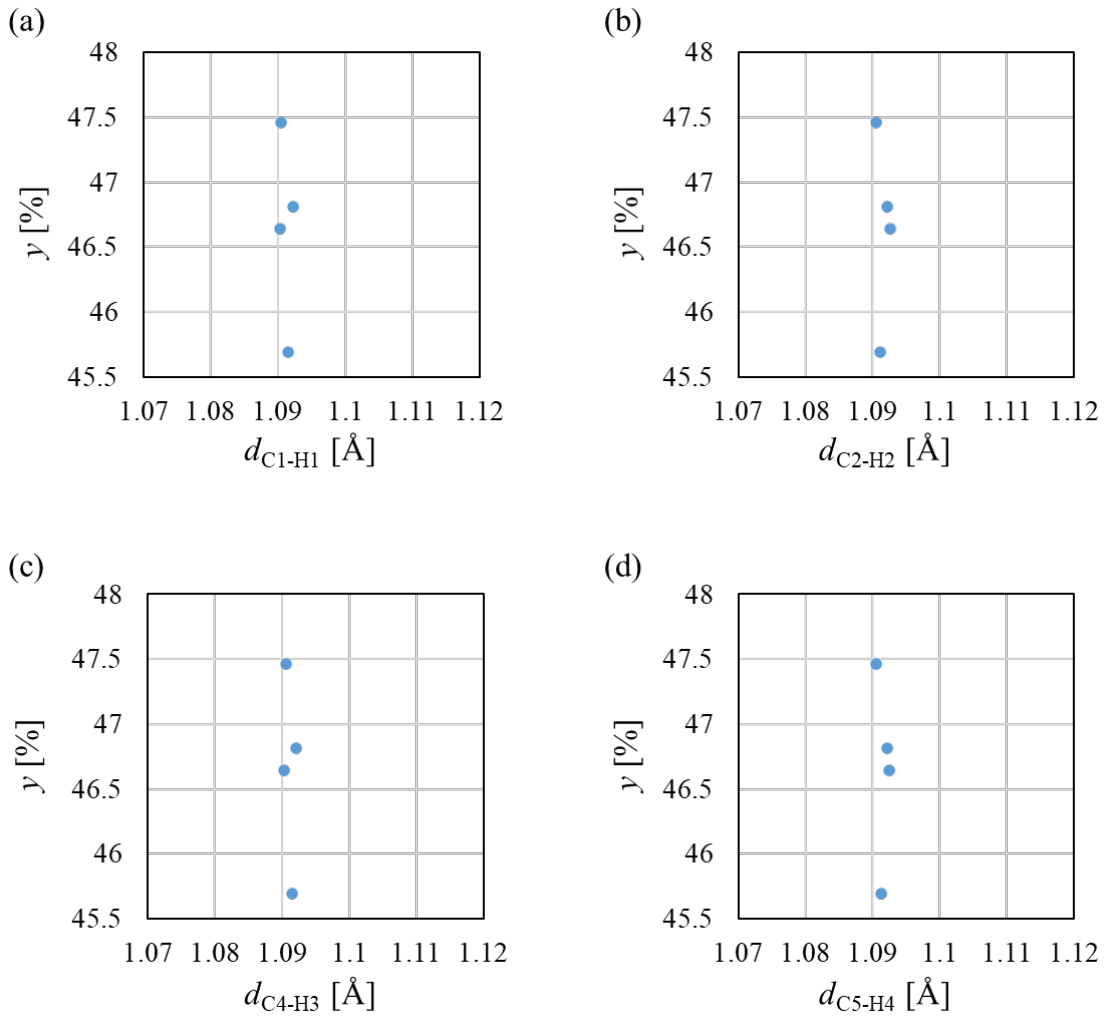

Figure S4. The plotted diradical character (*y*) against distances (a) $d_{C1\text{-}H1}$, (b) $d_{C2\text{-}H2}$, (c) $d_{C4\text{-}H3}$, and (d) $d_{C5\text{-}H4}$, where $d_{A\text{-}B}$ refers to the distance between atoms A and B atoms, and indices for C atoms are shown in the main text (Fig. 4(a)).

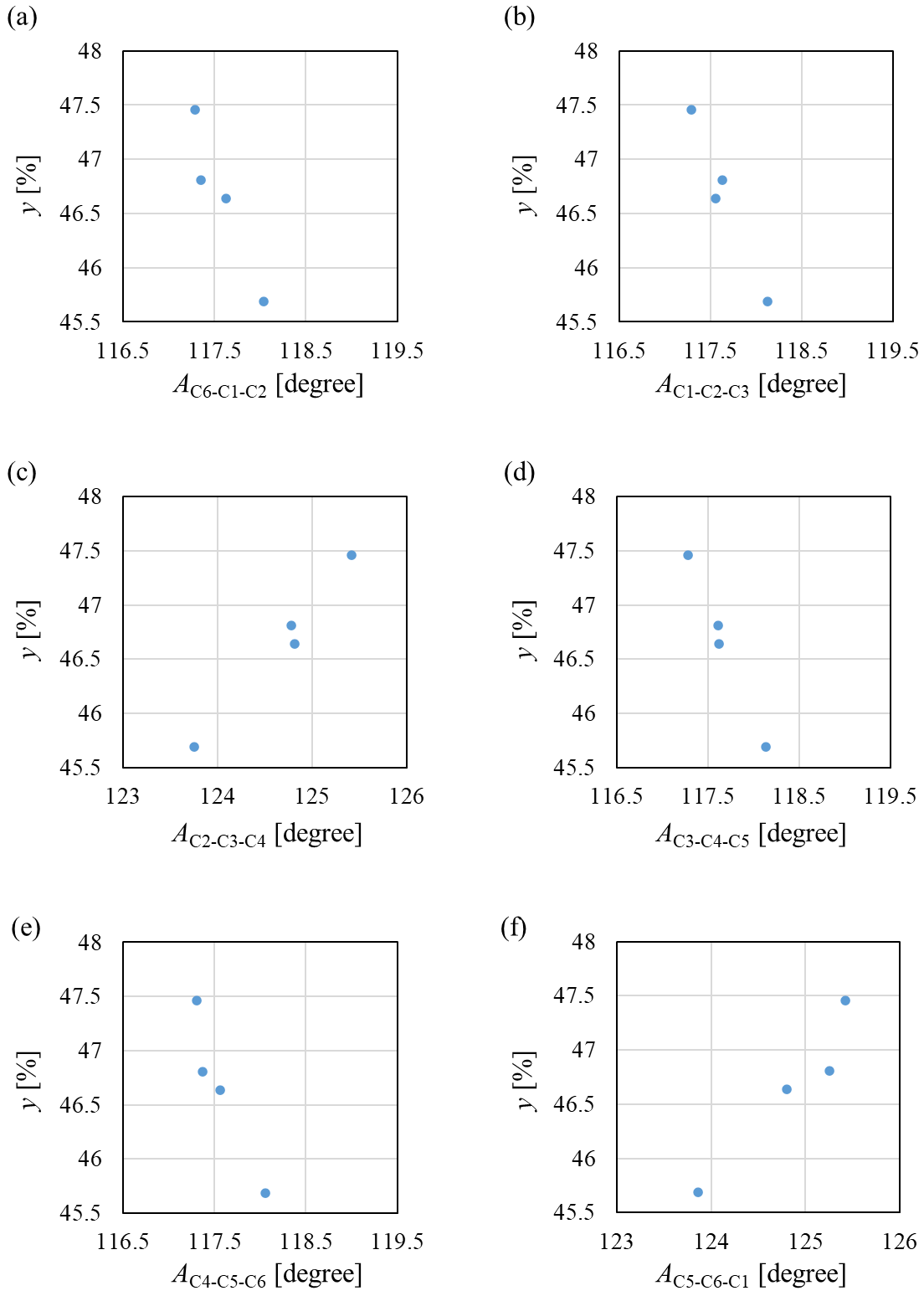

Figure S5. The plotted diradical character (y) against angles (a) $A_{C6\text{-}C1\text{-}C2}$, (b) $A_{C1\text{-}C2\text{-}C3}$, (c) $A_{C2\text{-}C3\text{-}C4}$, (d) $A_{C3\text{-}C4\text{-}C5}$, (e) $A_{C4\text{-}C5\text{-}C6}$, and (f) $A_{C5\text{-}C6\text{-}C1}$, where $A_{A\text{-}B\text{-}C}$ refers to the angle between atoms ABC (around atom B), and indices for C atoms are shown in the main text (Fig. 4(a)).

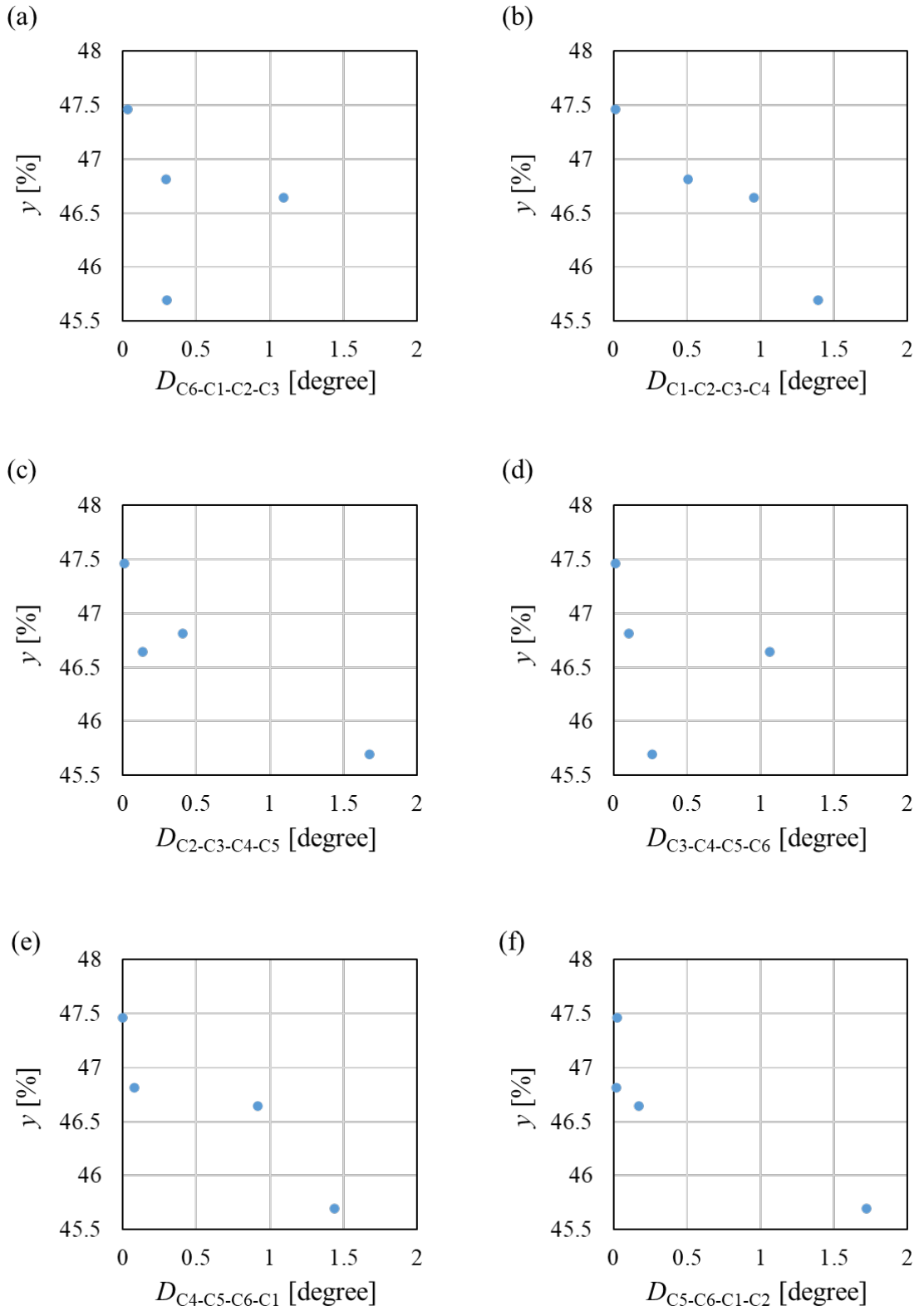

Figure S6. The plotted diradical character (*y*) against dihedral angles (a) $D_{C6\text{-}C1\text{-}C2\text{-}C3}$, (b) $D_{C1\text{-}C2\text{-}C3\text{-}C4}$, (c) $D_{C2\text{-}C3\text{-}C4\text{-}C5}$, (d) $D_{C3\text{-}C4\text{-}C5\text{-}C6}$, (e) $D_{C4\text{-}C5\text{-}C6\text{-}C1}$, and (f) $D_{C5\text{-}C6\text{-}C1\text{-}C2}$, where $D_{A\text{-}B\text{-}C\text{-}D}$ refers to the angles between planes ABC and BCD, and indices for C atoms are shown in the main text (Fig. 4(a)).

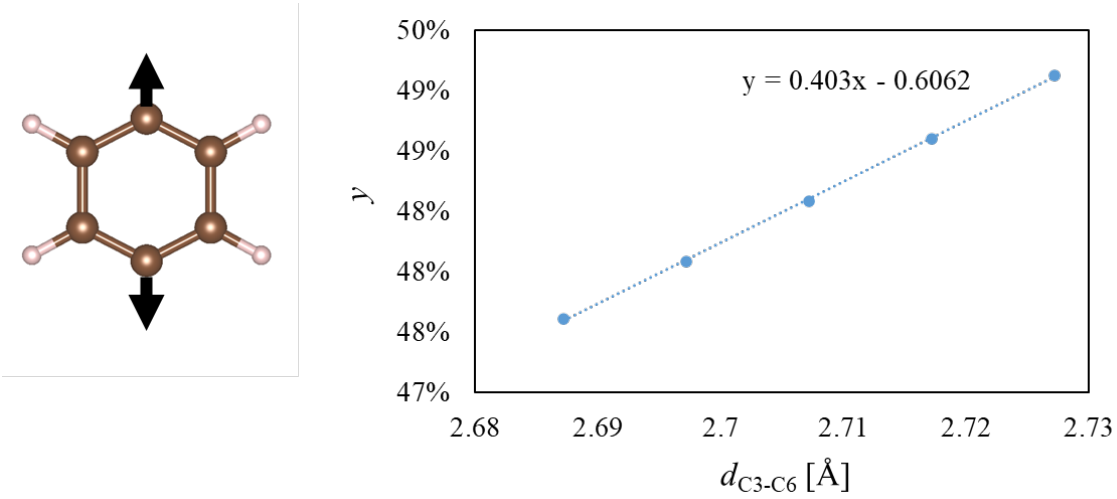

Figure S7. The dependence of the diradical character on the distance between atoms C3 and C6 when the optimised p-benzyne is systematically distorted (the distortion is shown in the left panel). The explanation for this index is provided in the main text (Fig. 4(a)).

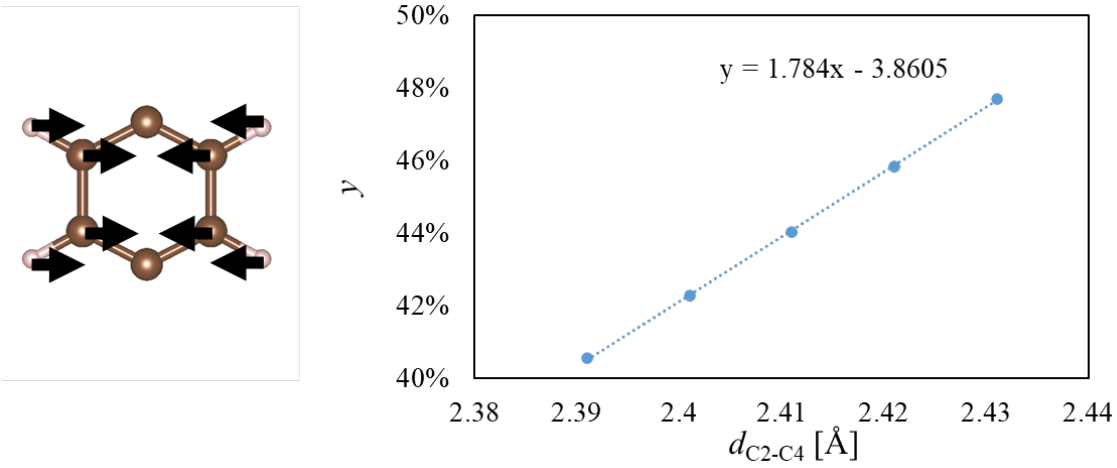

Figure S8. The dependence of the diradical character on the distance between atoms C2 and C4 atoms when the optimised p-benzyne is systematically distorted (the distortion is shown in the left panel). The explanation for this index is provided in the main text (Fig. 4(a)).

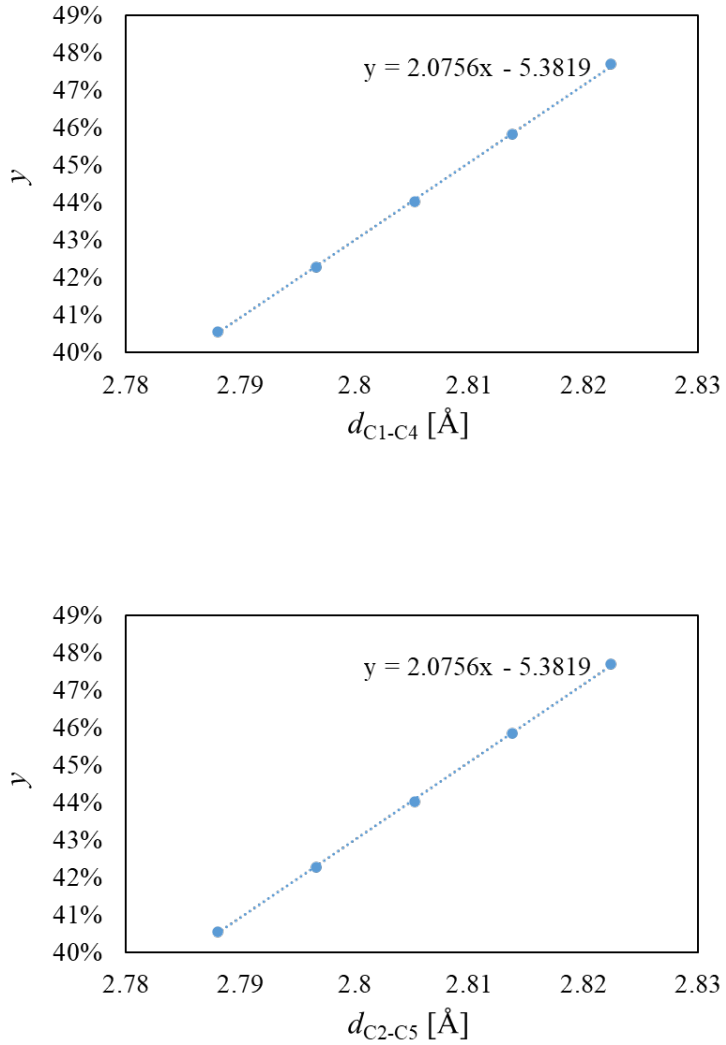

Figure S9. The dependence of the diradical character on the distance between atoms (a) C1 and C4 and (b) C2 and C5 when the optimised p-benzyne is systematically distorted (the distortion is shown in Fig. S8). The explanation for this index is provided in the main text (Fig. 4(a)).

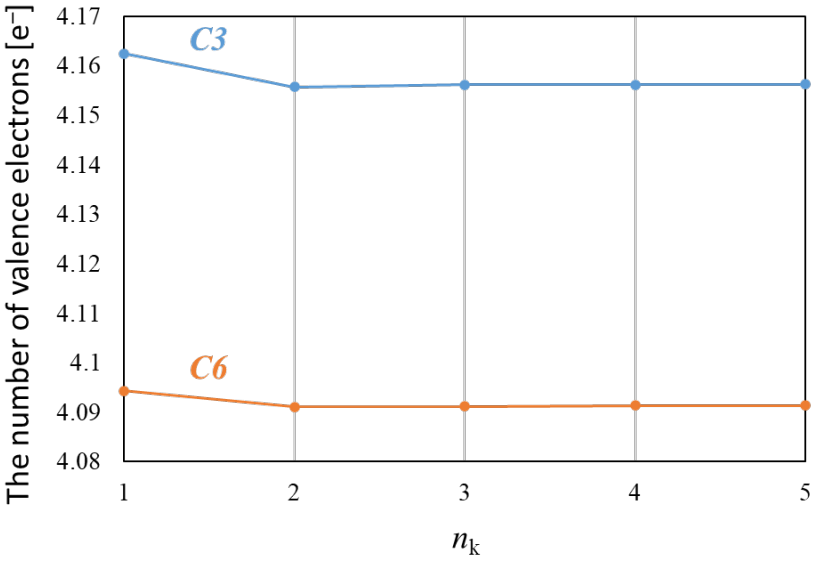

Figure S10. The dependences of the number of valence electrons of C3 and C6 in the Model D on the k-points sampling ($n_k \times n_k \times 1$).