Supplementary Information

Insights into the Catalytic Mechanism of Chlorophenol 4-Monooxygenase: A Quantum Mechanics/Molecular Mechanics Study

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Keywords

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Six pages

Contains one Table and three Figures

Table S1 Dependency of the energy barriers on the QM size, the smaller QM region (98 atoms) contains oxidation state of co-enzyme FADHOOH, functional groups of residues Arg100, Phe153, Val154, Thr192, His289 and the substrate 2,4,5-TCP, the larger QM region (212 atoms) contains additional residues such as Ala104, Pro150, Leu151, Glu251, Phe285, Cal288, Arg295, Arg366, and Phe442.

Figure S1 The optimized structures at B3LYP/6-31G(d,p) level and calculated energies at B3LYP/6-311++G(d,p) level by using Gaussian program. Energies are given in kcal mol^{-1}. A and C are two possible conformations of 5-Cl-2-H-BQ while B and D are two possible conformations of 4-Cl-3-H-BQ (P-10).

Figure S2 Optimized reactant (R'-1), transition state (TS'-2), and intermediate (IM'-3) structures for the hydroxylation step of TftD towards 2,4,6-TCP at B3LYP/6-31G(d,p)//CHARMM22 level. The unit of the bond distances and imaginary frequency are in Å and cm^{-1}.

Figure S3 Optimized reactant (R-6), transition state (TS-7), and intermediate (IM-8) structures for the hydroxylation step of TftD towards 2,5-DiCHQ at B3LYP/6-31G(d,p)//CHARMM22 level. The unit of the bond distances and imaginary frequency are in Å and cm^{-1}.

Table S1 Dependency of the energy barriers on the QM size, the smaller QM- region (98 atoms) contains oxidation state of co-enzyme FADHOOH, functional groups of residues Arg100, Phe153, Val154, Thr192, His289 and the substrate 2,4,5-TCP, the larger QM-region (212 atoms) contains additional residues such as Ala104, Pro150, Leu151, Glu251, Phe285, Cal288, Arg295, Arg366, and Phe442.

Snapshots	Energy barriers/(kcal mol^{-1})	
	Smaller QM-region	Larger QM-region
3 ns	16.3	15.2
4 ns	18.2	17.4
5 ns	24.7	23.2

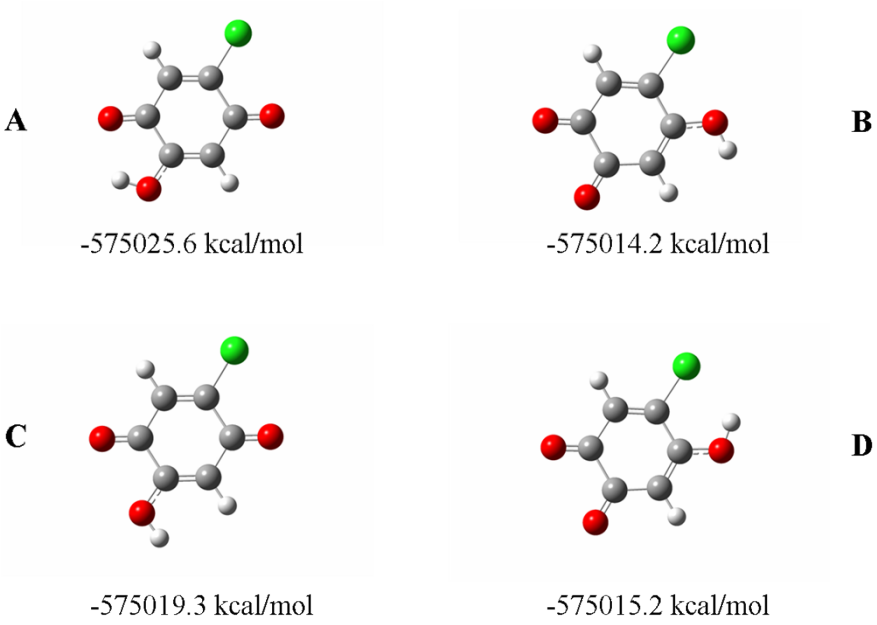
A
-575025.6 kcal/mol
B
-575014.2 kcal/mol
C
-575019.3 kcal/mol
D
-575015.2 kcal/mol

Figure S1

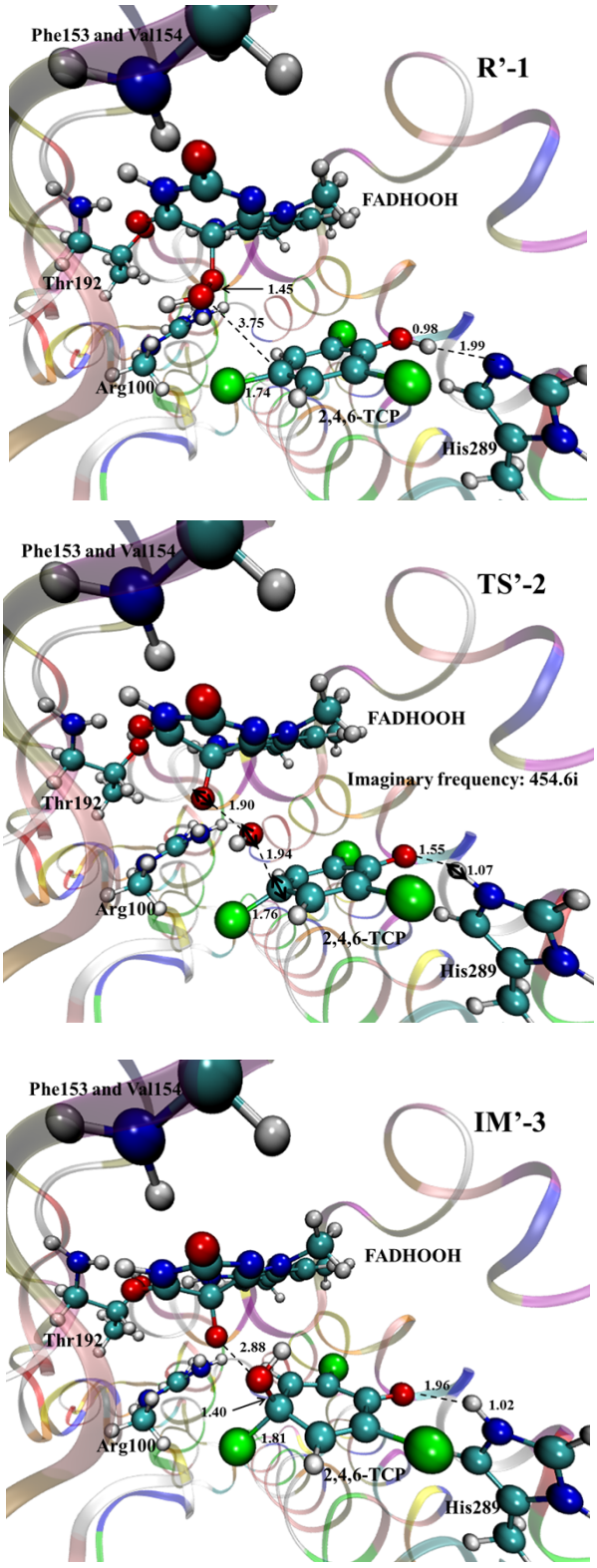
Phe153 and Val154
R'-1
FADHOOH
Thr192
1.45
3.75
0.98
1.99
Arg100
1.74
2,4,6-TCP
His289
Phe153 and Val154
TS'-2
FADHOOH
Imaginary frequency: 454.6i
Thr192
1.90
1.94
1.55
1.07
Arg100
1.76
2,4,6-TCP
His289
Phe153 and Val154
IM'-3
FADHOOH
Thr192
2.88
1.96
1.02
1.40
1.81
Arg100
2,4,6-TCP
His289

Figure S2

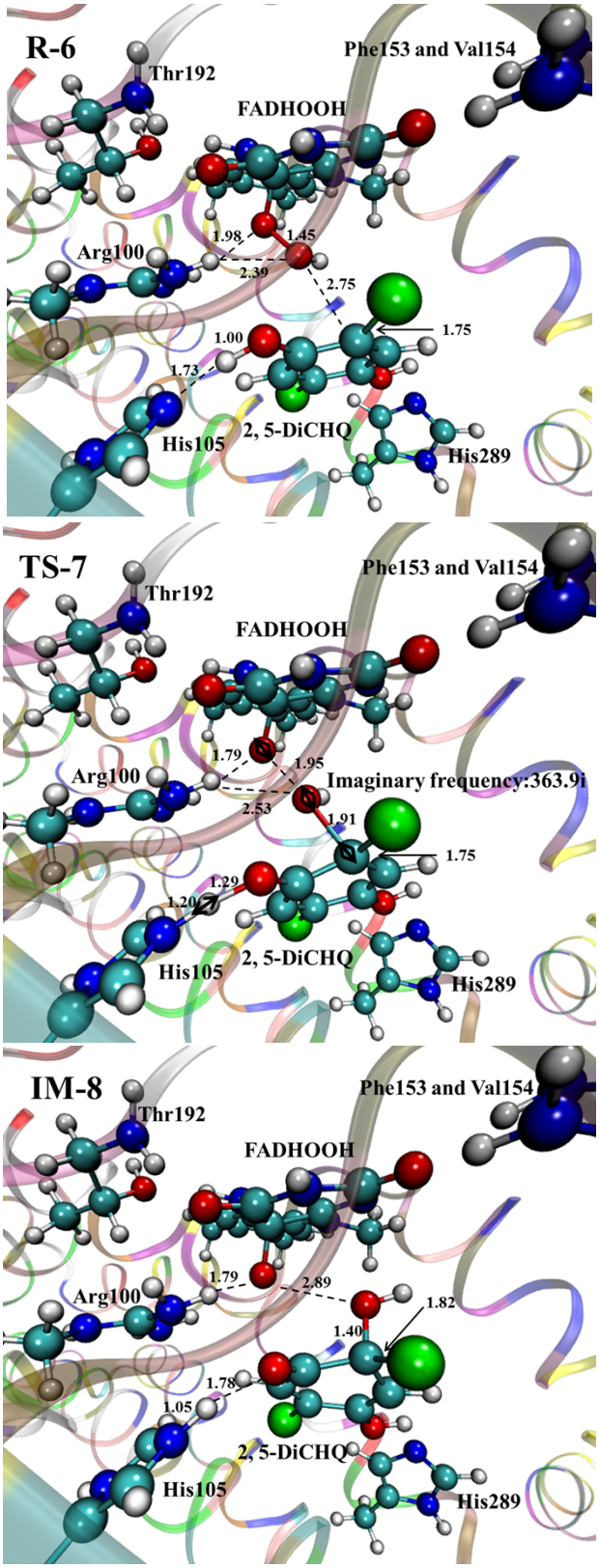
R-6
Thr192
Phe153 and Val154
FADHOOH
1.98
1.45
Arg100
2.39
2.75
1.75
1.00
1.73
His105
2, 5-DiCHQ
His289
TS-7
Thr192
Phe153 and Val154
FADHOOH
1.79
1.95
Arg100
Imaginary frequency:363.9i
2.53
1.91
1.75
1.29
1.20
His105
2, 5-DiCHQ
His289
IM-8
Thr192
Phe153 and Val154
FADHOOH
1.79
2.89
Arg100
1.82
1.40
1.78
1.05
2, 5-DiCHQ
His105
His289

Figure S3