Electronic supplementary information

Construction of Supramolecular Laccase Enzymes and Understanding of Catalytic Dye Degradation Using Multispectral and Molecular Docking Approaches

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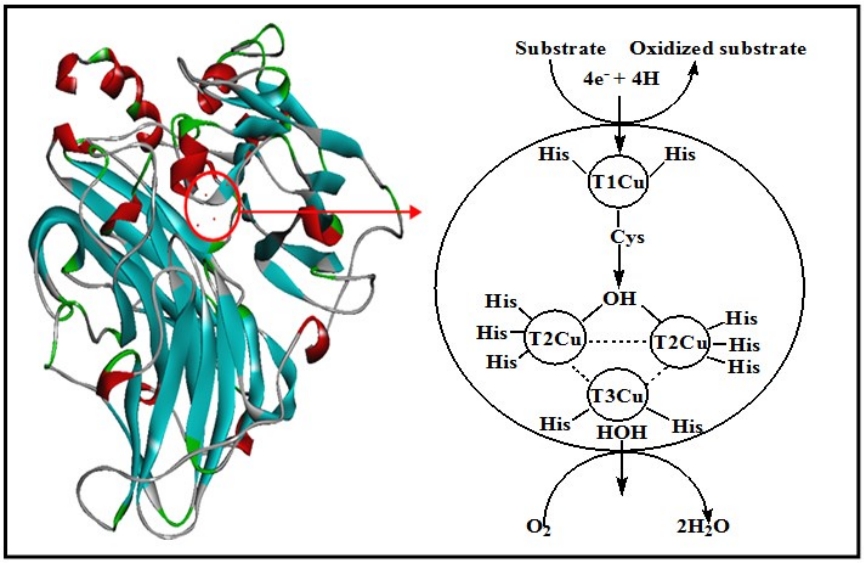

Fig. S1 Catalytic oxidation mechanism of the laccase active site

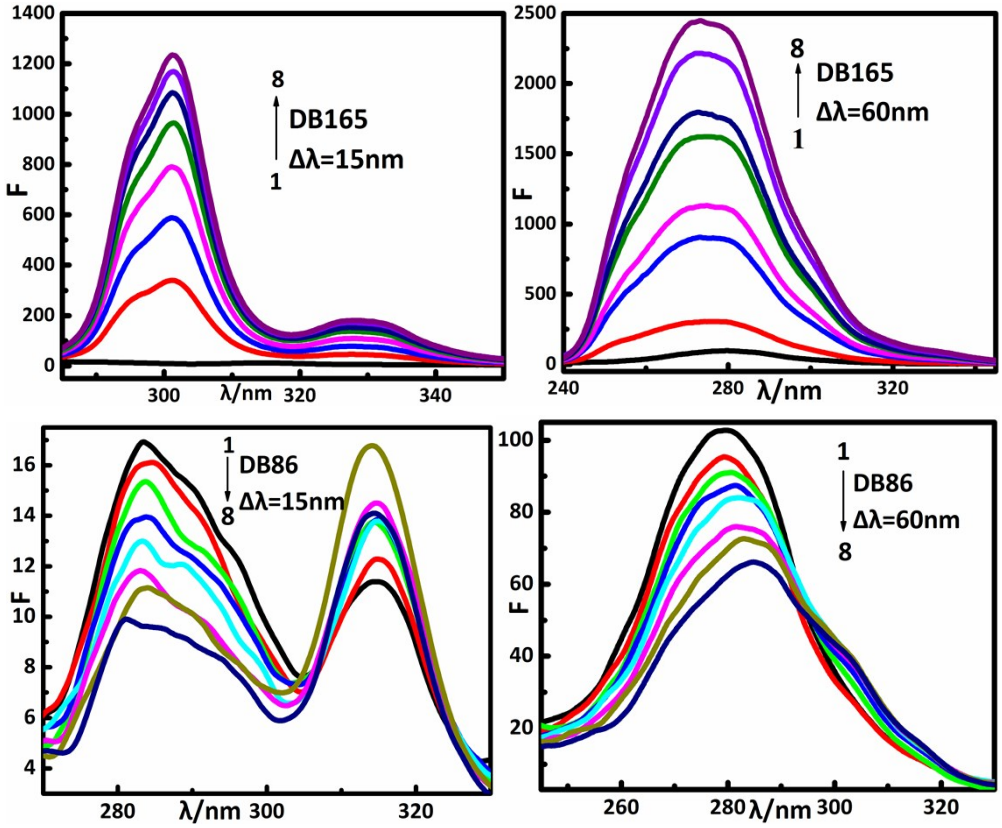

Fig. S2 DB165 and DB86 molecular synchronous fluorescence spectrum with SL-DSA (Δλ=15 nm, Δλ=60 nm), [$C_{Dyes} = C_{LAC} = 1.0\times10^{-5}$ M， T=298K， pH=4.5]

References

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[S2] D. Singh, K. K. Sharma, S. Jacob, S. K. Gakhar, Molecular Docking of Laccase Protein from Bacillus Safensis DSKK5 Isolated from Earthworm Gut: A Novel Method to Study Dye Decolorization Potential, *Water Air Soil Pollut*., 2014, **225**, 2175.

[S3] G. McMullan, C. Meehan, A. Conneely, N. Kirby, T. Robinson, P. Nigam, I. M. Banat, R. Merchant, W. F. Smyth, Microbial decolorization and degradation of textile dyes. *Appl. Microbiol. Biotechnol*. 2001, **56**, 81-87.

[S4] M. Chivukula, V. Renganathan, Phenolic azo dye oxidation by laccase from Pyricularia oryzae. *Appl. Environ. Microbiol*., 1995, **61**, 4347-4377.

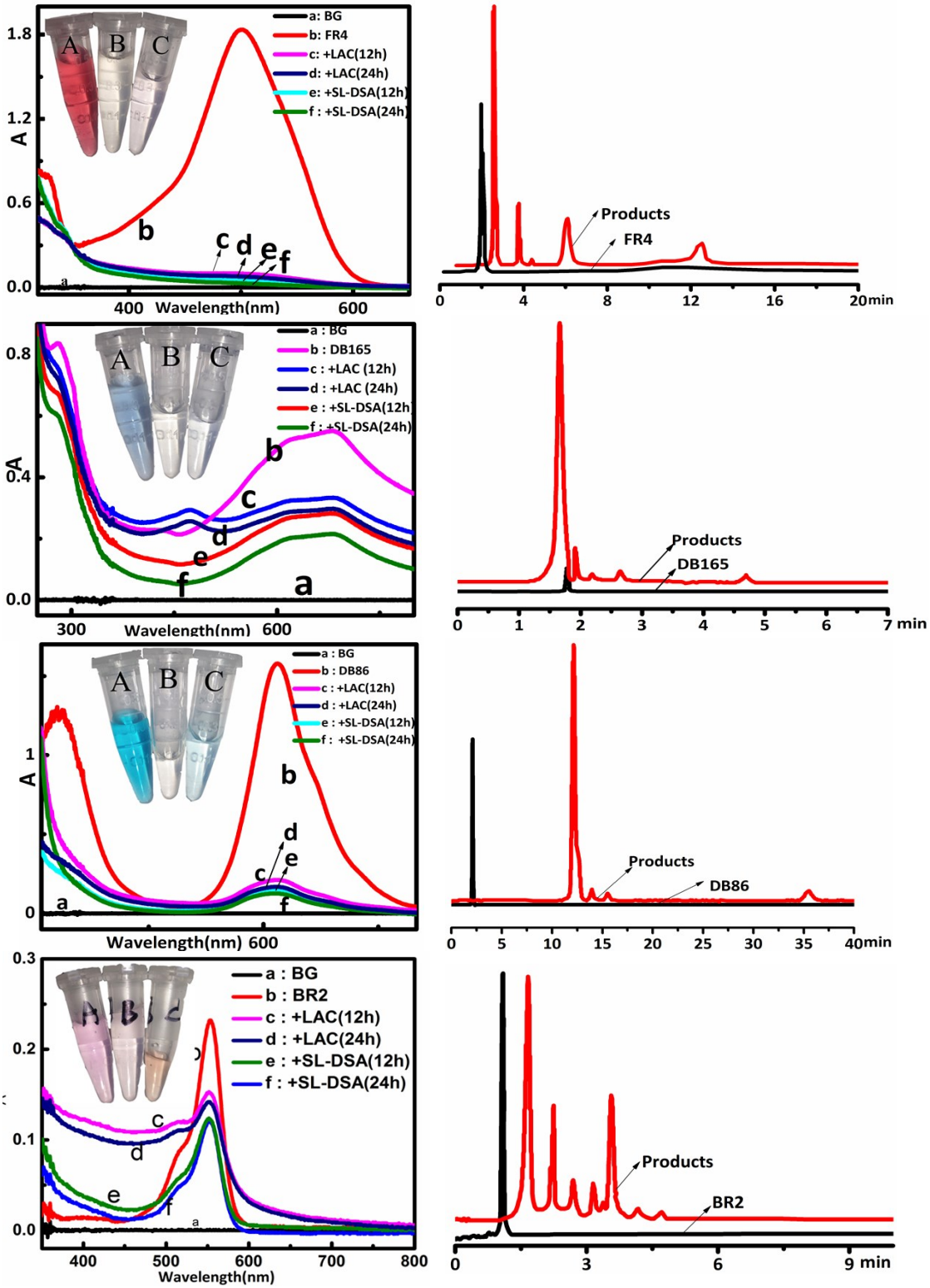
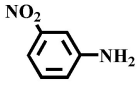
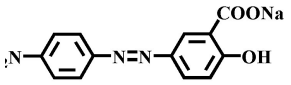
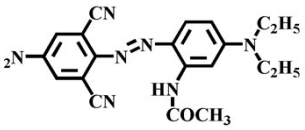
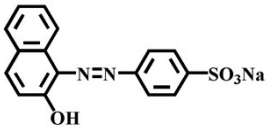
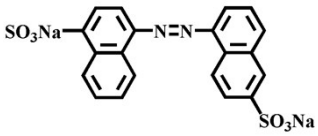
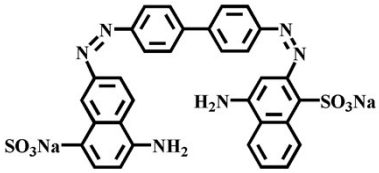
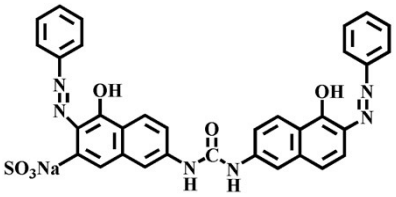


Fig. S3 Comparison of chromaticity, UV spectrum and HPLC before and after decolorization of dyes

Table S1 Chemical information and structural characteristics of the 13 kinds of dyes

Name of 13 dyes	Chemical characteristics	Category	Chemical structure of dyes
1. Azoic Diazo Component 7 (ADC 7), CAS: 99-09-2	MF: $C_6H_6N_2O_2$ MW: 138.12 g.mol^{-1} λ_{max}: 375 nm	Monoazo	
2. Mordant Orange1 (MO1) CAS: 2243-76-7	MF: $C_{13}H_8N_3NaO_5$ MW: 309.21 g.mol^{-1} λ_{max}: 385 nm	Monoazo	
3. Disperse Blue 165 (DB165), CAS: 41642-51-7	MF:$C_{20}H_{19}N_7O_3$ MW: 405.41 g.mol^{-1} λ_{max}: 694 nm	Monoazo	
4. Acid Orange 7 (AO7), CAS: 633-96-5	MF: $C_{16}H_{11}N_2NaO_4S$ MW: 350.33 g.mol^{-1} λ_{max}: 498 nm	Monoazo	
5. Food Red 4 (FR4), CAS: 2302-96-7	MF: $C_{20}H_{12}N_2Na_2O_7S_2$ MW: 502.43 g.mol^{-1} λ_{max}:500 nm	Monoazo	
6. Direct Red 28 (DR28),CAS: 573-58-0	MF: $C_{32}H_{22}N_6Na_2O_6S_2$ MW: 696.67 g.mol^{-1} λ_{max}: 497 nm	Bisazo	
7. Direct Orange 26(DO26), CAS: 3626-36-6	MF: $C_{33}H_{20}N_6Na_4O_{15}S_4$ MW: 960.55 g.mol^{-1} λ_{max}: 496 nm	Bisazo	

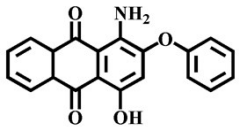
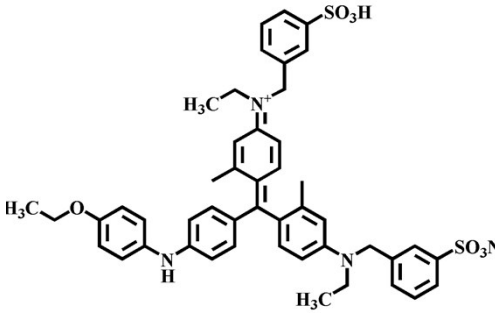
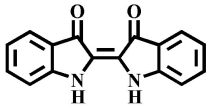
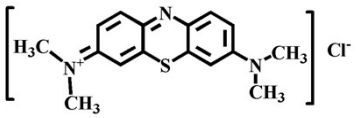
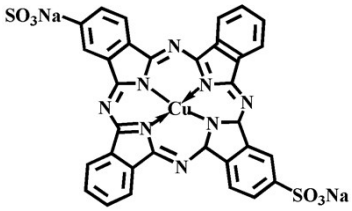
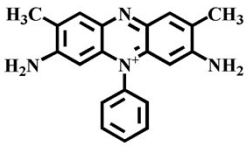
8. Disperse Red 60 (DR60), CAS: 12223-37-9	MF: $C_{20}H_{13}NO_4$ MW: 331.32 g.mol^{-1} λ_{max}: 520 nm	Anthraquinone	
9. Acid Blue 90 (AB90), CAS: 6104-58-1	MF: $C_{45}H_{44}N_3NaO_7S_2$ MW: 825.97 g.mol^{-1} λ_{max}: 595 nm	Triarylmethane	
10. Vat Blue1 (VB1), CAS: 482-89-3	MF: $C_{16}H_{10}N_2O_2$ MW: 262.26 g.mol^{-1} λ_{max}: 715 nm	Indigo	
11. Basic Blue 9 (BB9), CAS: 61-73-4	MF: $C_{16}H_{18}ClN_3S$ MW: 319.85 g.mol^{-1} λ_{max}: 664 nm	Sulfide dyes	
12. Direct Blue 86 (DB86), CAS: 1330-38-7	MF: $C_{34}H_{16}CuN_8Na_2O_6S_2$ MW: 982.18 g.mol^{-1} λ_{max}: 616 nm	Phthalocyanine	
13. Basic Red 2 (BR2), CAS: 477-73-6	MF: $C_{20}H_{19}ClN_4$ MW: 350.84g.mol^{-1} λ_{max}: 563 nm	Heterocyclic	

Table S2 Calculation results of combined distance

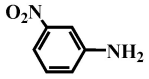
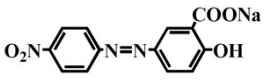
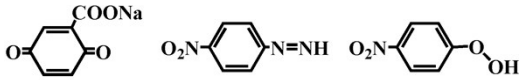
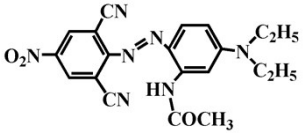
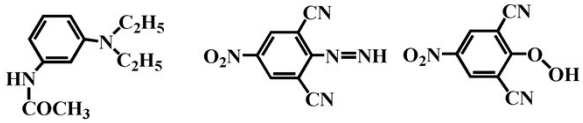
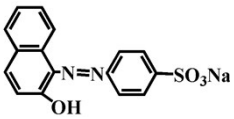
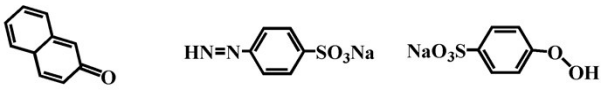
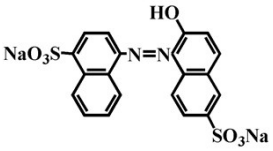
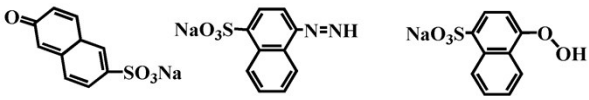
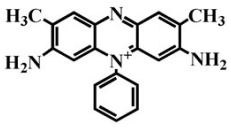
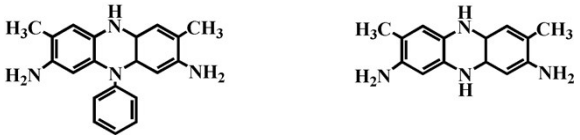
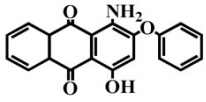
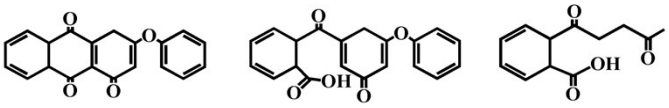
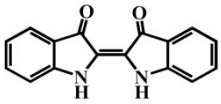
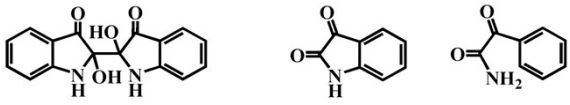
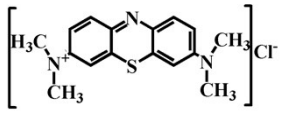
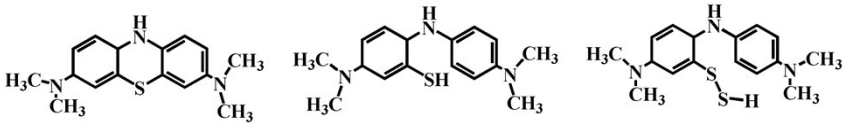
Dyes	J(cm^3·L / mol)	E	R_0/nm	*r*/nm
DB86	8.27×10^{-16}	0.6877	1.62	1.42
FR4	1.53×10^{-16}	0.6512	1.22	1.10
AB90	1.24×10^{-16}	0.5902	2.54	2.39
BB9	3.06×10^{-14}	0.7605	2.95	2.44
DO26	7.03×10^{-16}	0.4444	1.58	1.63

Table S3 The internal effective binding molecular model of the SL-DSA decolorization reaction system for the other 11 kinds of dyes

Dyes	Docking score kJ/mol	Amino acid residues that interact with chromophore	π-π stacking	Unfavorable
DR28	-79.48	Interaction between Asp206\Phe457 and the -N=N- bond and sulfosulfide atoms of DR28	Phe265\Phe457\Pro163 Pro394\Ala240\Pro396 Ile455 \Arg161\Ala461	Phe162\His458\ Phe239
AB90	-93.20	Interaction between Asp456\His111\ Phe344\Glu460 and the -C=N-\-C=C- bonds and sulfosulfide atoms of AB90	His454\Ala80\Phe344\Asp 456\Ser113\His398\His400 \Leu459 \His66	His64\Cu1501\ Cu1500\Pro346\ Thr114\Val82
DO26	-12.10	Interaction between Phe344\Ala80\ His111\Gly159\Ser343 and the -N=N- bond, sulfosulfide atoms of DO26	Glu460\ Val82\ Pro346\Arg157\ Ala156\Arg161	Ser113\Phe457\ Leu459
DR60	-8.03	Interaction between Phe344\Leu459\Gl y462 and the aniline group, phenolic hydroxyl group atom of DR60	Glu460\Pro346	Gly462\Arg161\ Arg157
FR4	-8.32	Interaction between Pro346\Arg161 and the -N=N- bond of FR4	Phe162\Pro391\Phe332	Nothing
ADC7	-5.79	Interaction between Leu459\Phe344 an d the benzene amino of ADC7	Pro346	Nothing
MO1	-7.25	Interaction between Glu460\Leu459\ Arg161 and the -N=N- bond of MO1	Leu459\Pro346\His111	Nothing
AO7	-7.20	Interaction between Glu460\Arg161 and the -N=N- bond of AO7	Thr345\Pro346\Phe344\ Arg157	Nothing
BB9	-5.80	Interaction of Phe265\Asp206 with aromatic ring -C–N- bond of BB9	Phe162	Nothing

VB1	-7.11	Interaction of Pro346\Glu460\Arg161 with the -C=N- bond of VB1	Leu459\Ala80\Ser113	Nothing
BR2	-4.66	Interaction of Phe162\Phe344\Glu460\ Pro346 with the -C=N- bond of BR2	Glu460\Arg161	Nothing

Table S4 The expected structure of the degradation products of the other 10 dyes

Dyes	Original dye structure	Prediction of degradation products
ADC7		
MO1		
DB165		
AO7		
FR4		
BR2		
DR60		
VB1		
BB9		

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