Chart S1. The calculated ground-state geometric parameters of the four dye molecules TC1, TC2, TC3 and TC4 in vacuum and methanol solvent, including the information about some important bond lengths, bond angles and dihedral angles.

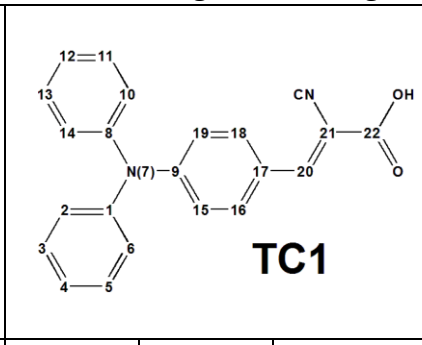
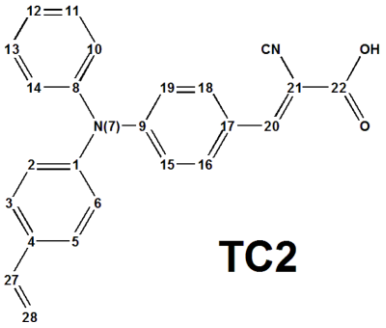
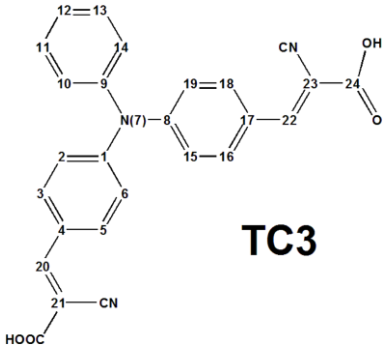
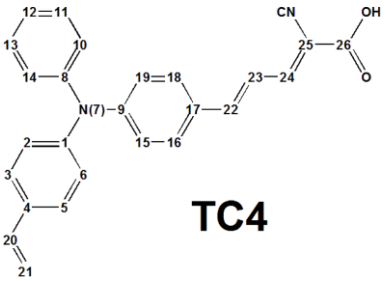
	 TC1			 TC2		
	Gas	Solvent	Solvent Shift	Gas	Solvent	Solvent Shift
R(1,7)	1.4249	1.4285	-0.0036	1.4222	1.4253	-0.0031
R(7,8)	1.4256	1.4285	-0.0029	1.4258	1.4287	-0.0029
R(7,9)	1.3949	1.3869	0.008	1.3961	1.3887	0.0074
R(17,20)	1.4468	1.4399	0.0069	1.4471	1.4405	0.0066
A(1,7,9)	121.0596	121.3209	-0.2613	121.1519	121.4196	-0.2677
A(8,7,9)	121.0322	121.3348	-0.3026	120.8426	121.1024	-0.2598
A(18,17,20)	125.5935	125.6782	-0.0847	125.58	125.6533	-0.0733
D(8,7,9,15)	153.7015	157.9777	-4.2762	153.0316	157.0951	-4.0635
D(1,7,9,19)	154.3001	158.4405	-4.1404	153.0948	156.7813	-3.6865
D(18,17,20,21)	0.4778	0.2357	0.2421	0.4618	0.9135	-0.4517
	 TC3			 TC4		
	Gas	Solvent	Solvent Shift	Gas	Solvent	Solvent Shift
R(1,7)	1.4057	1.4036	0.0021	1.4203	1.4219	-0.0016
R(7,8)	1.4071	1.404	0.0031	1.4237	1.4258	-0.0021
R(7,9)	1.4298	1.4327	-0.0029	1.4013	1.397	0.0043
R(17,22)	1.4515	1.4466	0.0049	1.4522	1.4479	0.0043
A(1,7,8)	121.9692	122.2945	-0.3253	120.9121	121.1793	-0.2672
A(8,7,9)	118.9735	118.8213	0.1522	120.6172	120.7579	-0.1407
A(18,17,22)	125.4424	125.4476	-0.0052	123.3983	123.5024	-0.1041
D(1,7,8,19)	146.8865	147.6876	-0.8011	149.8678	152.6244	-2.7566
D(9,7,8,15)	145.0045	145.7908	-0.7863	149.7443	152.3892	-2.6449
D(18,17,22,23)	1.4971	-0.2373	1.7344	1.0581	1.2866	-0.2285

Chart S2. The frontier molecular orbitals of the dye molecule TC4.

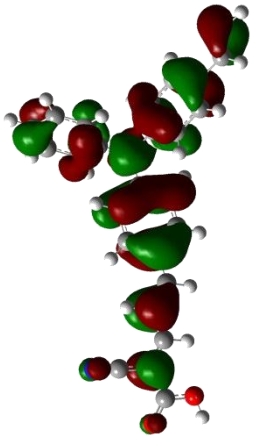
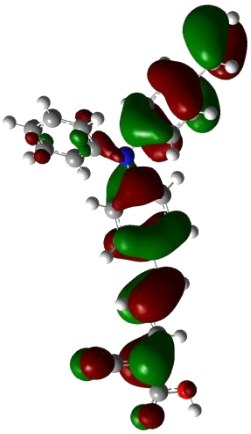
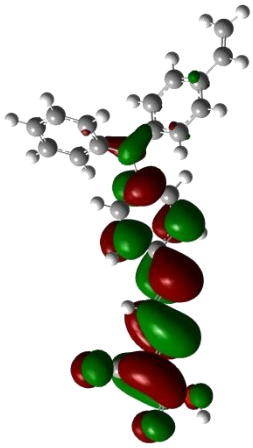
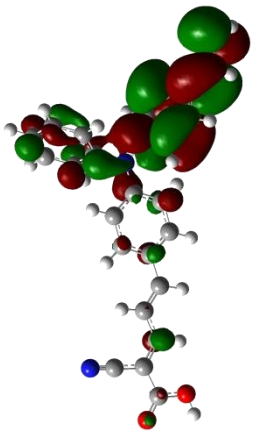
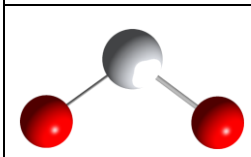
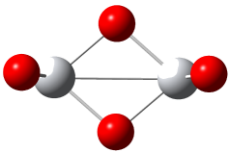
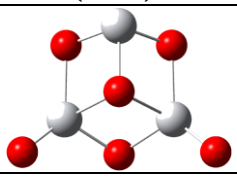
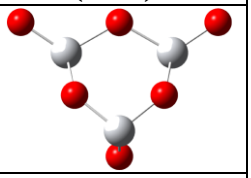
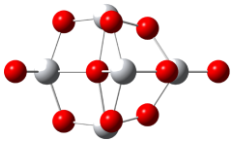
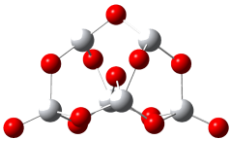
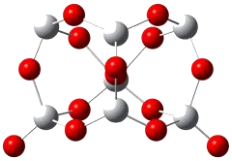
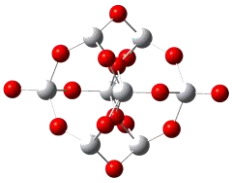
HOMO	HOMO-1	LUMO	LUMO+1
			

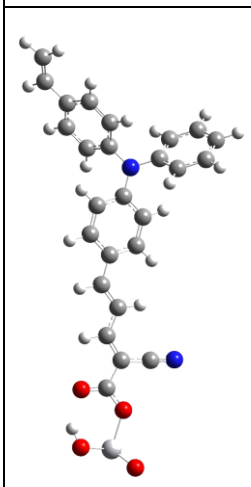
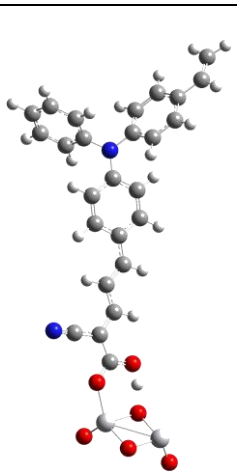
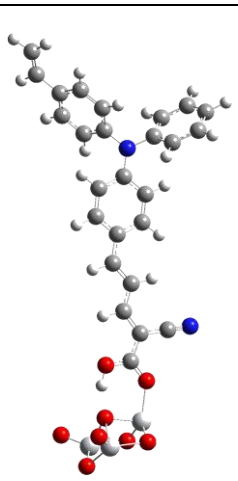
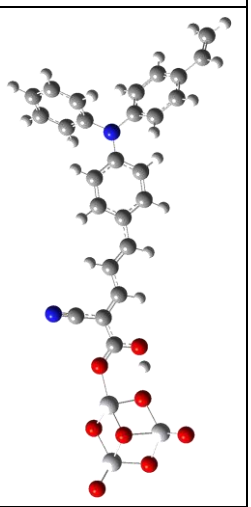
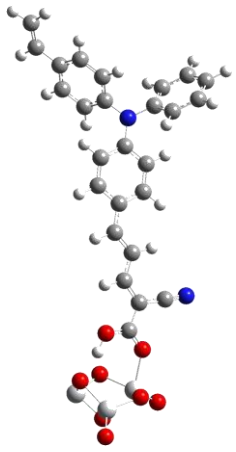
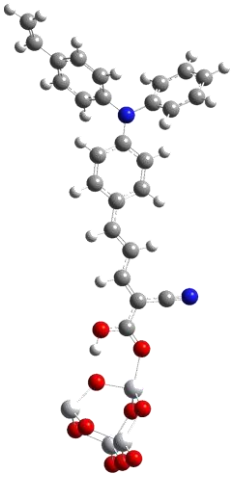
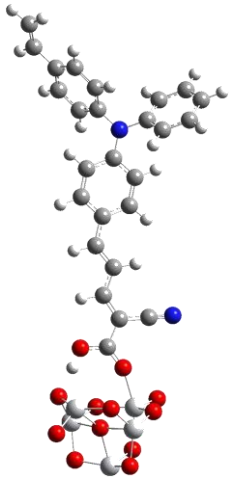
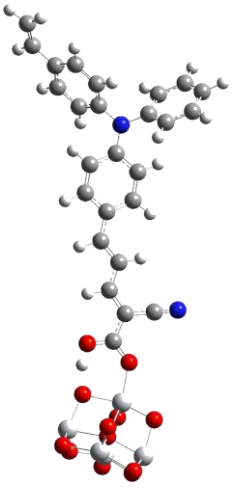
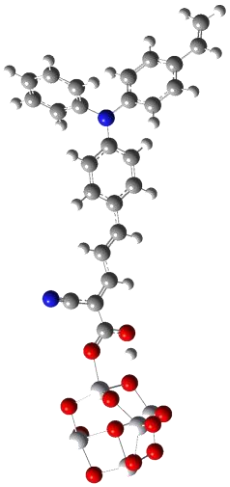
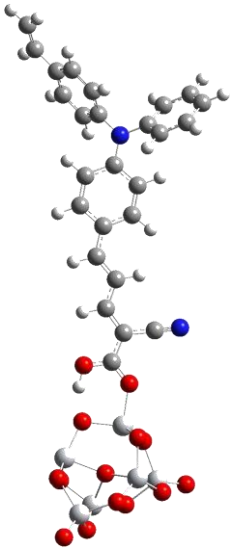
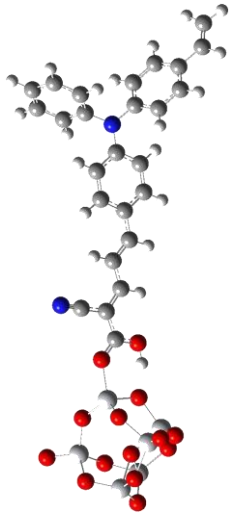
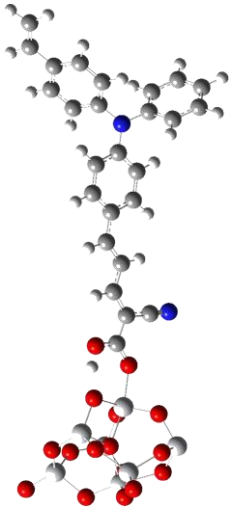
Chart S3. The calculated excitation energy E(eV), oscillator strength f, and the main configuration interaction (CI) coefficient for the S_1 excited states of the tested organic sensitizer/titanium dioxide cluster complex systems TC4-$(TiO_2)_n$ (n = 1~8). "a, b, c, d" respect the different binding for the sensitizer to 2, 3, 4, 5-fold-coordinated Ti of the clusters, respectively.

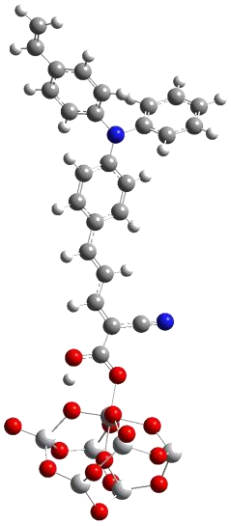
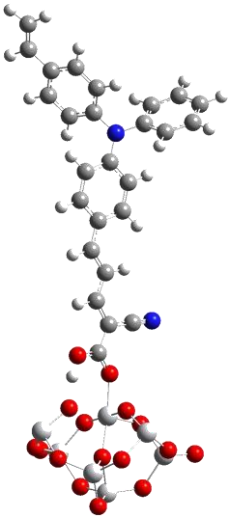
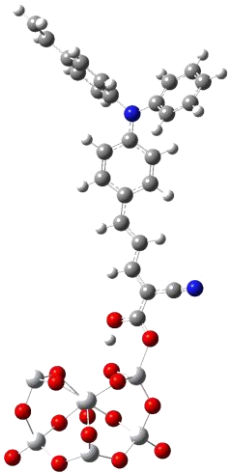
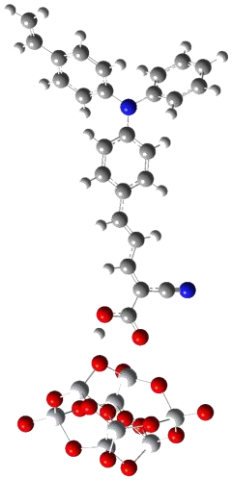
		E(eV, nm)	Main CI		f
TC4-TiO_2	1a	3.0038 (412.75)	H ->L	0.65223 (85%)	1.7264
TC4-$(TiO_2)_2$	1c	2.7888 (444.59)	H ->L	0.65881 (87%)	1.8252
TC4-$(TiO_2)_3$	1b	2.6488 (468.07)	H ->L	0.66172 (88%)	1.9643
	2b	2.6663 (465.00)	H ->L	0.66013 (87%)	2.0169
	3c	2.7640 (448.57)	H ->L	0.65624 (86%)	1.8316
TC4-$(TiO_2)_4$	1b	2.6655 (465.14)	H ->L	0.66205 (88%)	2.0793
TC4-$(TiO_2)_5$	1c	2.8189 (439.84)	H ->L	0.65654 (86%)	1.8959
	2c	2.6700 (464.36)	H ->L	0.65486 (86%)	2.0766
	3c	2.7074 (457.94)	H ->L	0.65533 (86%)	2.0090
TC4-$(TiO_2)_6$	1b	2.5761 (481.28)	H ->L	0.65119 (85%)	2.1594
	2c	2.5937 (478.02)	H ->L	0.66067 (87%)	2.0940
TC4-$(TiO_2)_7$	1c	2.6902 (460.88)	H ->L	0.63310 (80%)	2.0695
	2d	2.7345 (453.40)	H ->L	0.64666 (84%)	2.0184
	3d	2.7025 (458.77)	H ->L	0.64930 (84%)	1.9692
	4b	2.6841 (461.92)	H ->L	0.65734 (86%)	2.0939
TC4-$(TiO_2)_8$	1d	2.8133 (440.71)	H ->L	0.62743 (79%)	2.0323
	2c	2.6982 (459.51)	H ->L	0.63670 (81%)	1.7183
	3d	2.7607 (449.10)	H ->L	0.63386 (80%)	1.9597

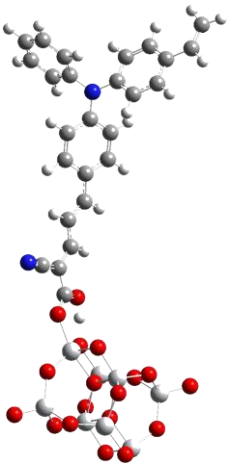
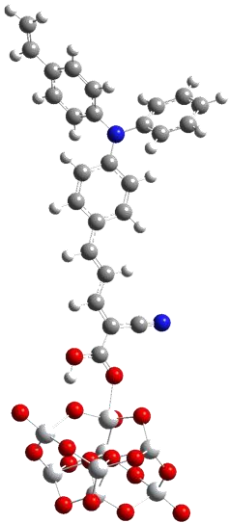
Figure S1. The optimized structures of (A) the titanium dioxide clusters and (B) the sensitizer/titanium dioxide cluster complex systems TC4-$(TiO_2)_n$ (n = 1~8). “a, b, c, d” respect the different binding for the sensitizer to 2, 3, 4, 5-fold-coordinated Ti of the clusters, respectively.

TiO2	$(TiO_2)_2$	$(TiO_2)_3$	$(TiO_2)_4$
			
$(TiO_2)_5$	$(TiO_2)_6$	$(TiO_2)_7$	$(TiO_2)_8$
			

(A)

TC4-(TiO_2)	TC4-(TiO_2)$_2$	TC4-(TiO_2)$_3$	
1a	1c	1b	2b
			
TC4-(TiO_2)$_3$	TC4-(TiO_2)$_4$	TC4-(TiO_2)$_5$	
3c	1b	1c	2c
			
TC4-(TiO_2)$_5$	TC4-(TiO_2)$_6$		TC4-(TiO_2)$_7$
3c	1b	2c	1c
			

TC4-$(TiO_2)_7$			TC4-$(TiO_2)_8$
2d	3d	4b	1d
			

TC4-$(TiO_2)_8$	
2c	3d
	

(B)