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.

	12=11 13 14-8 N 2=1 3 6	(7)-9 15-16 T	сл он 21-22 0 СП	$ \begin{array}{c} 12 = 11 \\ 13 \\ 14 \\ 14 \\ 19 = 18 \\ 19 = 18 \\ 17 \\ 20 \\ 0 \\ 15 \\ 15 \\ 15 \\ TC2 \end{array} $		
	45			27		
	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
	12 = 13 11 10 = 9 N(7) = 8 19 = 18 10 = 23 = 24 10 = 18 10 = 23 = 24 10 = 10 10 =			13 $12=11$ CN OH 13 $14-8$ $19=18$ $23-24$ O		
	2 1 15-16 3 4 5 20 21 CN HOOC					
	а 20 21 — С Ноос	13—16	ТСЗ	2	`15−16 [″] TC	4
	а 20 21 ноос Gas	Solvent	TC3 Solvent Shift	20 20 21 Gas	15-16 TC Solvent	4 Solvent Shift
R(1,7)	а 20 21 ноос Gas 1.4057	Solvent 1.4036	TC3 Solvent Shift 0.0021	2 20 21 Gas 1.4203	15-16 TC Solvent 1.4219	54 Solvent Shift -0.0016
R(1,7) R(7,8)	з 20 21-с ноос Gas 1.4057 1.4071	Solvent 1.4036	TC3 Solvent Shift 0.0021 0.0031	2 2 2 2 2 2 2 1 3 4 5 5 2 0 2 1 2 1 2 1 5 2 1 5 2 1 5 2 1 5 2 1 5 2 1 5 5 2 1 5 5 5 5	Solvent 1.4219 1.4258	Solvent Shift -0.0016 -0.0021
R(1,7) R(7,8) R(7,9)	з 20 21 4 3 20 21 20 21 4 4 5 20 21 4 6 5 3 4 4 5 5 20 21 6 5 5 5 1.4057 1.4057 1.4071 1.4298	Solvent 1.4036 1.404 1.4327	TC3 Solvent Shift 0.0021 0.0031 -0.0029	2 2 2 2 2 2 2 2 2 2 2 2 2 2	Solvent 1.4219 1.4258 1.397	Solvent Shift -0.0016 -0.0021 0.0043
R(1,7) R(7,8) R(7,9) R(17,22)	з 20 21 ноос Gas 1.4057 1.4071 1.4298 1.4515	Solvent 1.4036 1.404 1.4327 1.4466	TC3 Solvent Shift 0.0021 0.0031 -0.0029 0.0049	Gas 1.4203 1.4237 1.4013 1.4522	Solvent 1.4219 1.4258 1.397 1.4479	S olvent Shift -0.0016 -0.0021 0.0043 0.0043
R(1,7) R(7,8) R(7,9) R(17,22) A(1,7,8)	а 20 21 20 21 20 21 20 21 20 21 20 21 20 21 20 21 20 21 21 20 21 21 20 21 21 20 21 21 20 21 20 21 20 21 20 21 20 21 20 21 20 21 20 21 20 21 20 20 21 20 20 20 20 20 20 20 20 20 20	Solvent 1.4036 1.404 1.4327 1.4466 122.2945	TC3 Solvent Shift 0.0021 0.0031 -0.0029 0.0049 -0.3253	2 2 2 2 2 2 2 2 2 2 2 2 2 2	Solvent 1.4219 1.4258 1.397 1.4479 121.1793	Solvent Shift -0.0016 -0.0021 0.0043 0.0043 -0.2672
R(1,7) R(7,8) R(7,9) R(17,22) A(1,7,8) A(8,7,9)	а 3 20 21 4 20 21 4 21 4 5 20 21 4 5 20 21 4 5 5 1.4057 1.4071 1.4071 1.4298 1.4515 121.9692 118.9735	Solvent 1.4036 1.404 1.4327 1.4466 122.2945 118.8213	TC3 Solvent Shift 0.0021 0.0031 -0.0029 0.0049 -0.3253 0.1522	Gas 1.4203 1.4237 1.4013 1.4522 120.9121 120.6172	Solvent 1.4219 1.4258 1.397 1.4479 121.1793 120.7579	S olvent Shift -0.0016 -0.0021 0.0043 0.0043 -0.2672 -0.1407
R(1,7) R(7,8) R(7,9) R(17,22) A(1,7,8) A(1,7,8) A(18,17,22)	а 20 21 20 21 20 21 20 21 20 21 4057 1.4057 1.4057 1.4071 1.4298 1.4515 121.9692 118.9735 125.4424	Solvent 1.4036 1.404 1.4327 1.4466 122.2945 118.8213 125.4476	TC3 Solvent Shift 0.0021 0.0031 -0.0029 0.0049 -0.3253 0.1522 -0.0052	2 2 2 2 2 2 2 2 2 2 2 2 2 2	Solvent 1.4219 1.4258 1.397 1.4479 121.1793 120.7579 123.5024	Solvent Shift -0.0016 -0.0021 0.0043 0.0043 -0.2672 -0.1407 -0.1041
R(1,7) R(7,8) R(7,9) R(17,22) A(17,22) A(17,7,8) A(18,17,22) D(1,7,8,19)	а 20 20 21 20 21 20 21 20 21 20 21 21 21 20 21 21 21 21 21 21 21 21 21 21	Solvent 1.4036 1.404 1.4327 1.4466 122.2945 118.8213 125.4476 147.6876	TC3 Solvent Shift 0.0021 0.0031 -0.0029 0.0049 -0.3253 0.1522 -0.0052 -0.8011	Gas 1.4203 1.4237 1.4013 1.4522 120.9121 120.6172 123.3983 149.8678	Solvent 1.4219 1.4258 1.397 1.4479 121.1793 120.7579 123.5024 152.6244	X olvent Shift -0.0016 -0.0021 0.0043 0.0043 -0.2672 -0.1407 -0.1041 -2.7566
R(1,7) R(7,8) R(7,9) R(17,22) A(1,7,8) A(18,17,22) D(1,7,8,19) D(9,7,8,15)	за 20 21 20 21 21 20 21 21 20 21 21 21 21 21 21 21 21 21 21	Solvent 1.4036 1.404 1.4327 1.4466 122.2945 118.8213 125.4476 147.6876 145.7908	TC3 Solvent Shift 0.0021 0.0021 0.0029 0.0049 -0.3253 0.1522 -0.0052 -0.8011 -0.7863	2 2 2 2 2 2 2 2 2 2 2 2 2 2	Solvent 1.4219 1.4258 1.397 1.4479 121.1793 120.7579 123.5024 152.6244 152.3892	Solvent Shift -0.0016 -0.0021 0.0043 0.0043 -0.2672 -0.1407 -0.1041 -2.7566 -2.6449

HOMO	HOMO-1	LUMO	LUMO+1

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

Chart S3. The calculated excitation energy E(eV), oscillator strength f, and the main configuration interaction (CI) coefficient for the S₁ excited states of the tested organic sensitizer/titanium dioxide cluster complex systems TC4-(TiO₂)_n ($n = 1 \sim 8$). "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 $TC4-(TiO_2)_n$ ($n = 1 \sim 8$). "a, b, c, d" respect the different binding for the sensitizer to 2, 3, 4, 5-fold-coordinated Ti of the clusters, respectively.





