

## Supporting Materials

**Table S1.** Selected bond lengths ( $\text{\AA}$ ) and dihedral angles( $^{\circ}$ ) of R and Q in phase of vacuum and solvent.

Dyes	V-R	S-R	V-Q	S-Q
C2-C3 ( $\text{\AA}$ )	1.473	1.474	1.482	1.470
C4-O5 ( $\text{\AA}$ )	1.370	1.373	1.550	1.373
O5-C6 ( $\text{\AA}$ )	1.445	1.445	1.450	1.455
C1-C2-C3-C4 ( $^{\circ}$ )	-35.438	-35.780	-65.395	21.705
C3-C4-O5-C6 ( $^{\circ}$ )	128.985	129.893	54.764	116.200
C4-O5-C6-O7 ( $^{\circ}$ )	-120.791	-111.489	42.702	118.087

**Table S2.** The calculated PDOS of S-Q and S-R.

Dyes	S-Q	S-R
HOMO	97%	95%
LUMO	100%	99%

**Table S3.** The calculated NBO of S-Q and S-R.

Dyes	$q_{s0}$	$q_{s1}$	$\Delta_q$	$\Delta_q/q_{s0}$
S-Q	Black	0.34366	0.33281	-0.01085
	Red	-0.34362	-0.33283	0.01079
S-R	Black	1.36283	1.40278	0.03995
	Red	-1.36282	-1.40274	-0.03992

**Table S4.** The dipole moments  $\mu$  (D), the polarizability  $\alpha$  (a.u.), the average polarizability  $\alpha_{\text{tot}}$  (esu), the anisotropy of the polarizability  $\Delta \alpha$  (esu), and the first hyperpolarizability  $\beta_{\text{tot}}$  (esu) of S-Q and S-R.

Dyes	S-Q	S-R
$\mu_x$	2.369	-4.851
$\mu_{\text{normal}}$	15.997	2.001
$\mu_z$	3.178	-1.538
$A_{xx}$	465.123	614.275
$A_{xy}$	39.469	20.355
$A_{yy}$	445.354	648.377
$A_{xz}$	-16.362	-68.116
$A_{yz}$	21.555	32.774
$A_{zz}$	179.191	399.567
$\beta_{xxx}$	-669.782	1665.694
$\beta_{xxy}$	-2999.843	-595.604
$\beta_{xyy}$	-3003.958	-2069.564
$\beta_{xxz}$	-70.385	-884.451
$\beta_{xyz}$	-128.747	-155.406
$\beta_{yyz}$	6.126	366.560
$\beta_{xzz}$	83.683	367.315
$\beta_{yzz}$	-22.914	203.916
$\beta_{zzz}$	75.598	-386.774

$\mu_{tot}$	16.480	5.468
$\alpha_{tot}$	363.222	554.073
$\Delta_\alpha$	288.732	270.128
$\beta_{tot}$	5639.846	3619.623

**Table S5.** Charge transfer distance (DCT), overlap extent (Sr), t and H index derived through electron and hole analysis.

Dyes	D <sub>CT</sub> /Å	Sr	t/Å	H/Å
S-Q	0.370	0.6935	-1.808	3.289
S-R	0.650	0.70478	-1.247	3.211

**Table S6.** The experimental stokes shift of Q and R in three different solvents (water, dimethyl sulfoxide (DMSO), methanol).

Experiment	Absorption(nm)		Emission(nm)		Stokes(nm)	
	Solvent	Q	R	Q	R	Q
Water	347	350	396.6	467.4	49.6	117.4
DMSO	358	357	398.6	438.8	79.6	81.8
Methanol	349	353	430	458.4	81	105.4

**Table S7.** The theoretical calculated stokes shift of Q and R.

Theory	Absorption (nm)		Emission (nm)		Stokes (nm)	
	Solvent	Q	R	Q	R	Q
Water	347.48	336.43	417.01	422.63	69.53	86.2
DMSO	348.03	336.91	418.94	423.98	70.91	87.07
Methanol	347.44	336.26	418.80	422.18	71.36	85.92

**Table S8.** The fluorescent lifetime of S-Q and S-R.

Dyes	E (eV)	$\lambda$ (nm)	f	$\tau$ (ns)
S-Q	1.815	683.11	0.0324	215.92
S-R	2.985	415.41	0.4773	5.42

**Table S9.** The I-V parallel experiments of S-Q and S-R.

Dyes	S-Q		S-R	
$J_{sc}$ (mAcm <sup>2</sup> )	5.293	5.480	5.346	1.826
$V_{oc}$ (V)	0.596	0.582	0.585	0.547
$\eta$ (%)	2.008	2.151	2.156	0.713
				0.685

**Figure captures:**

**Fig S1.** The schematic structure of DSSC devices

**Fig S2.** The optimized ground state structures of S-Q and S-R.

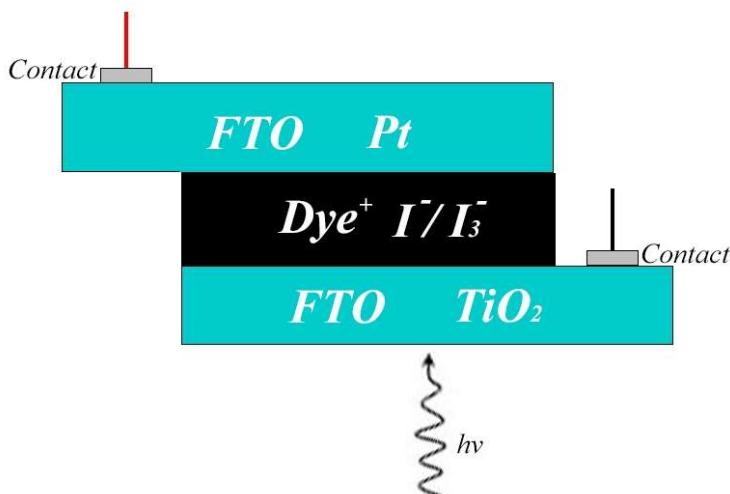
**Fig S3.** The labels of S-Q and S-R corresponding to the FT-IR spectrum.

**Fig S4.** Frontier molecular orbitals of S-Q and S-R.

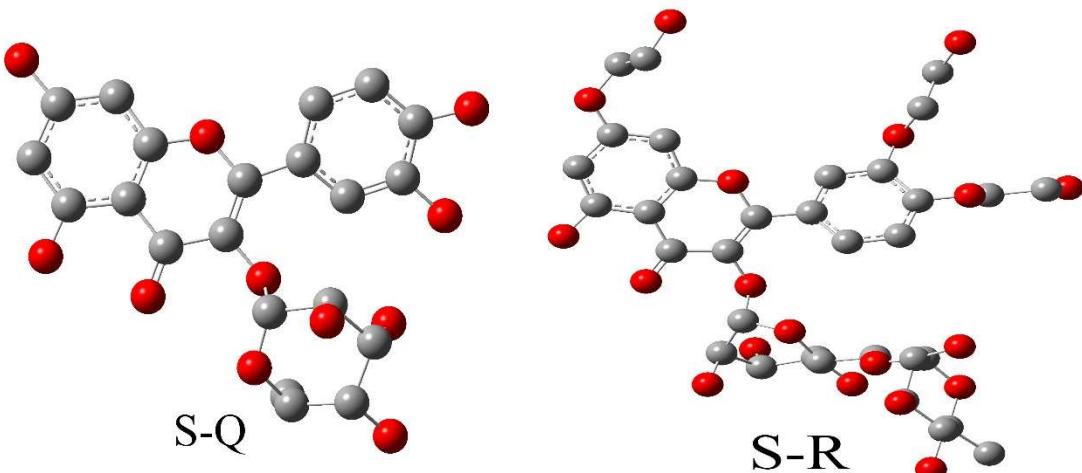
**Fig S5.** The CDD pictures of S-Q and S-R.

**Fig S6.** The composition of Q (A, B, C represents the group of 3,5,7-trihydroxy-4H-chromen-4-one; pyrocatechol; 2-methyl-tetrahydro-2H-pyran-3,4,5-triol, respectively) and R (A,B,C represents the group of 2,2'-(1,2-Phenylenebis(oxy)diethanol;3,5-dihydroxy-7-(2-hydroxyethoxy)-2-methyl-4H-chromen-4-one;2-methyl-6-((3,4,5-trihydroxytetrahydro-2H-pyran-2-yl)methoxy)tetrahydro-2-H-pyran-3,4,5-triol respectively).

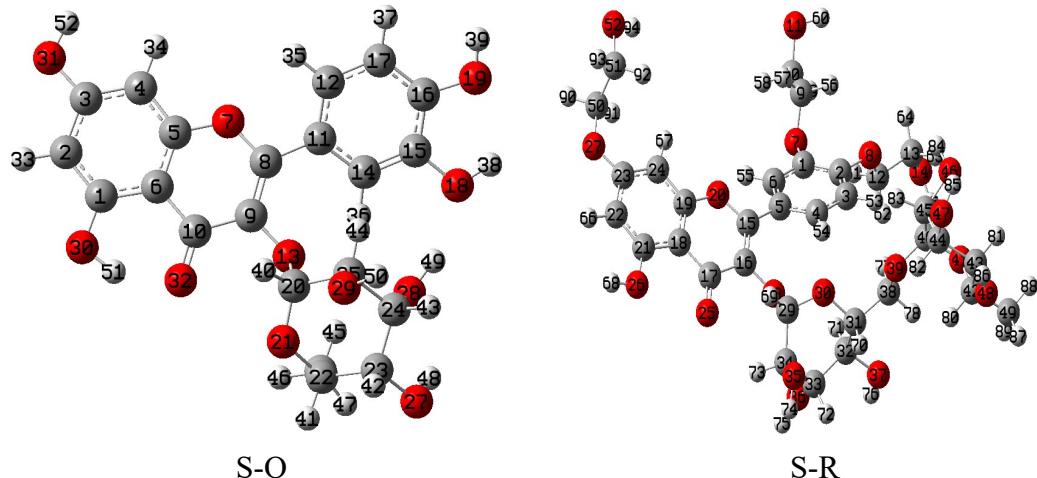
### Dye-sensitized Solar Cells



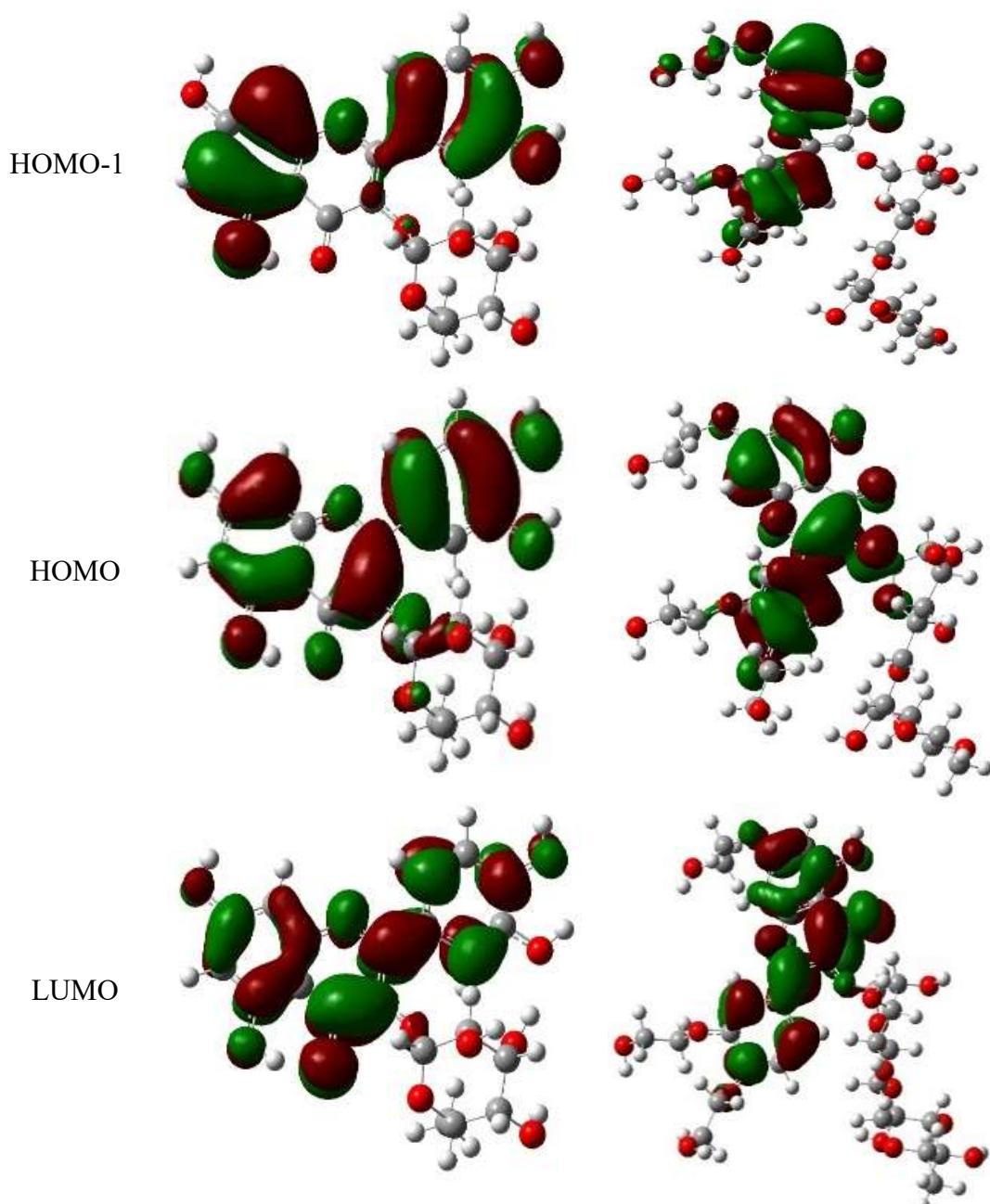
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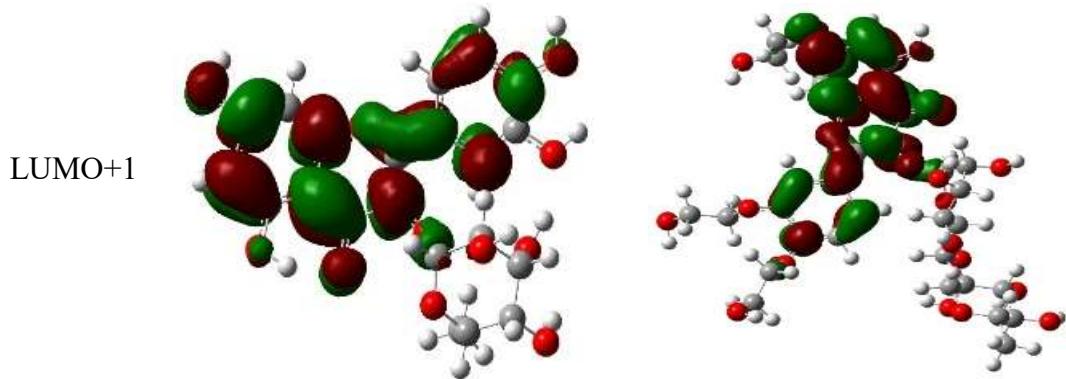


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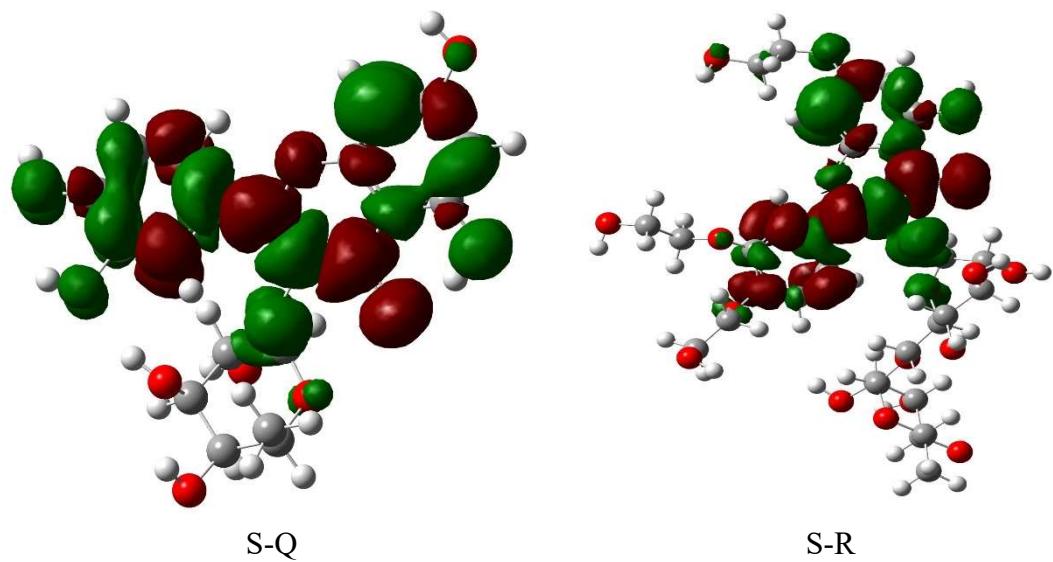


**Fig S3.** The labels of V-Q and V-R corresponding to the FT-IR spectrum.

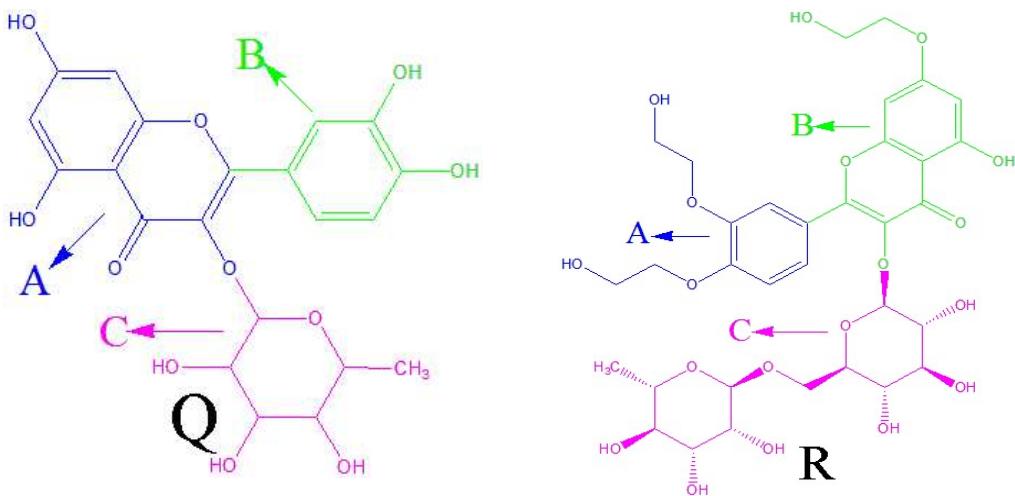




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