Figure S1. (a) UV−vis absorption spectra of sensitizers JM203/JM203+CDCA and JM204/JM204+CDCA attached on 4μm TiO_2 film immerged in CH_3CN solutions with 0.1 M $LiClO_4$. (b) Photograph of dye adsorption on 6μm TiO_2.
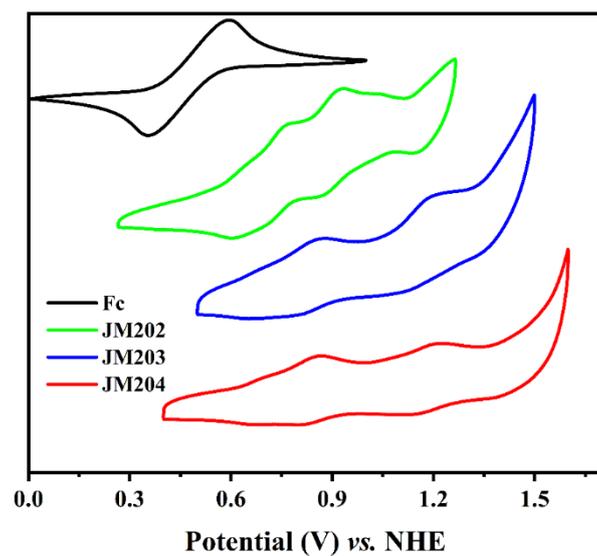
Figure S2. Cyclic voltammograms of **JM202**, **JM203** and **JM204** sensitizers

	JM202
LUMO+1	
LUMO	
HOMO	
HOMO-1	
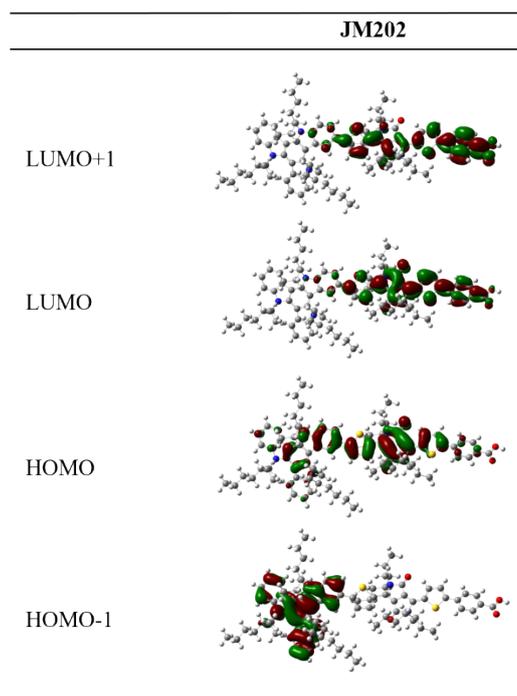
Figure S3. The electron distribution in HOMO, HOMO-1 and LUMO, LUMO+1

levels of JM202 by DFT calculations..

26.94°

JM202

54.02° **JM203**

54.79° **JM204**
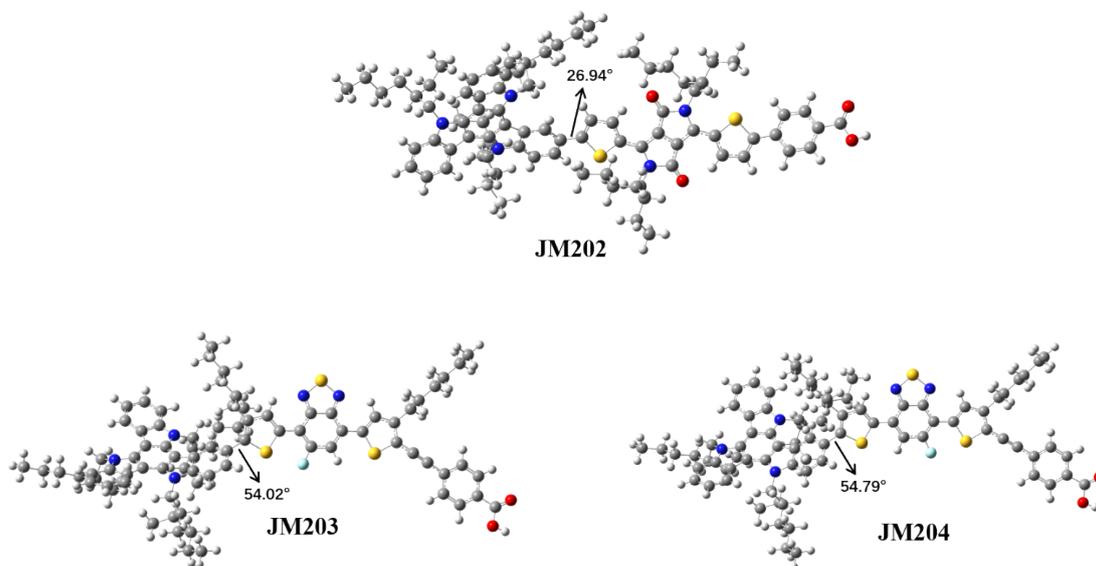
Figure S4. Dihedral angles between the neighboring units calculated from optimized

structures.
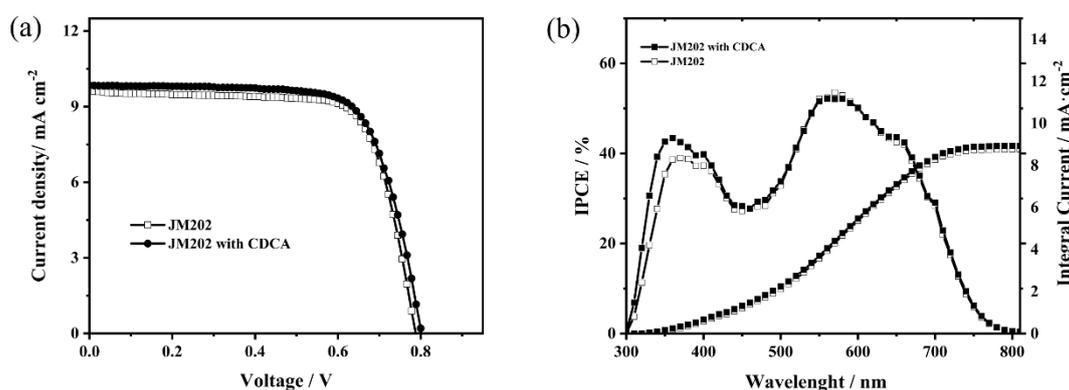
Figure S5. (a) Current−voltage (J−V) characteristics of the DSSCs measured under simulated AM1.5G. (b) IPCE spectra and integrated photocurrents of the DSSCs based on JM202.

Table S1. Photophysical and electrochemical properties of sensitizers on DFT calculations

Dye	$E_{0-0}^{DFT[a]}$ (eV)	$E_{H-1}^{DFT[b]}$ (eV)	$E_H^{DFT[b]}$ (eV)	$E_L^{DFT[b]}$ (eV)	$E_{L+1}^{DFT[b]}$ (eV)
JM202	2.14	-5.24	-4.98	-2.84	-1.90
JM203	2.07	-5.14	-5.07	-3.00	-2.16
JM204	2.18	-5.21	-5.14	-2.96	-2.08

[a] Energy gaps (E_{0-0}^{DFT}), [b]H, H-1 and L, L+1 represent HOMO, HOMO-1 and LUMO, LUMO+1 respectively. The frontier orbital energy levels (E_H^{DFT} and EL^{DFT}) with respect to vacuum are calculated at the B3LYP/6-311G(d,p) level of theory for dye in vacuum, $\lambda_{abs\ max}^{DFT}$ is derived from DFT calculation at the B3LYP/6-311G(d,p) level of theory for dye in vacuum, Energy gaps (DFT) are calculated via $E_{0-0}^{DFT} = E_L^{DFT} - E_H^{DFT}$.
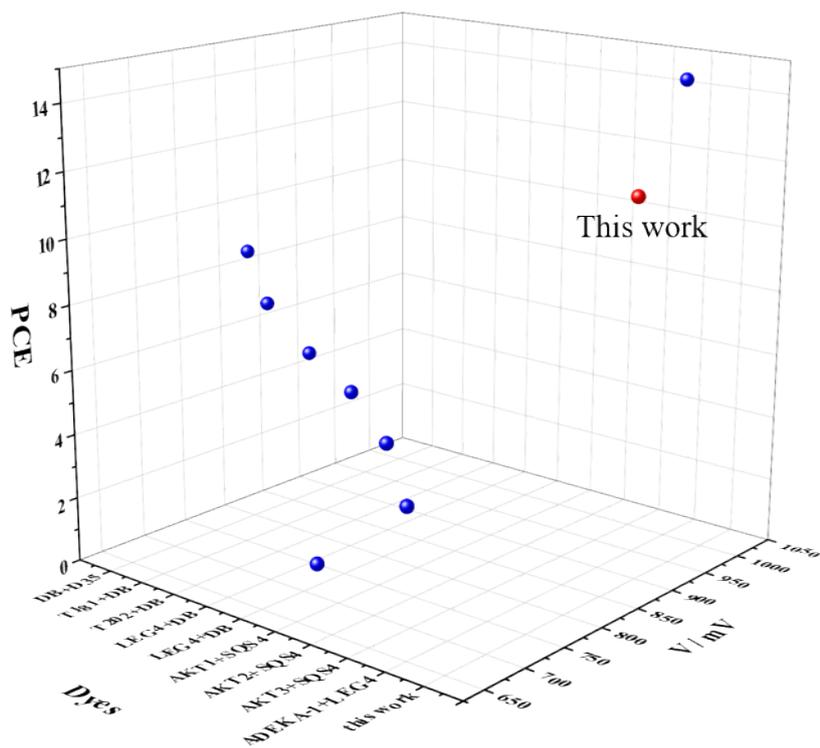
Figure S6. Comparison of reported and current work on cobalt electrolyte-based pure organic photosensitizers for co-sensitization.

Table S2. Optimization data of DSSCs devices based on JM202-JM204 and co-sensitization under different conditions.

Dye	Electrolyte	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF (%)	PCE (%)
JM202	[Cu(tmby)$_2$]$^{2+/+}$	0.82	8.61	70.76	5.0
JM203	[Cu(tmby)$_2$]$^{2+/+}$	0.96	5.19	75.91	3.8
JM204	[Cu(tmby)$_2$]$^{2+/+}$	0.99	8.57	75.00	6.3
JM202:JM203 (60 μM:40μM)	[Co(bpy)$_3$]$^{3+/2+}$	0.82	13.04	68.81	7.4
JM202:JM203 (50 μM:50μM)	[Co(bpy)$_3$]$^{3+/2+}$	0.83	14.93	67.63	8.5
JM202:JM203 (40 μM:60μM)	[Co(bpy)$_3$]$^{3+/2+}$	0.84	13.95	66.58	7.8
JM202:JM204 (60 μM:40μM)	[Co(bpy)$_3$]$^{3+/2+}$	0.86	17.88	69.15	10.7
JM202:JM204 (50 μM:50μM)	[Co(bpy)$_3$]$^{3+/2+}$	0.89	20.38	64.24	11.7
JM202:JM204 (40 μM:60μM)	[Co(bpy)$_3$]$^{3+/2+}$	0.89	18.39	66.08	10.8
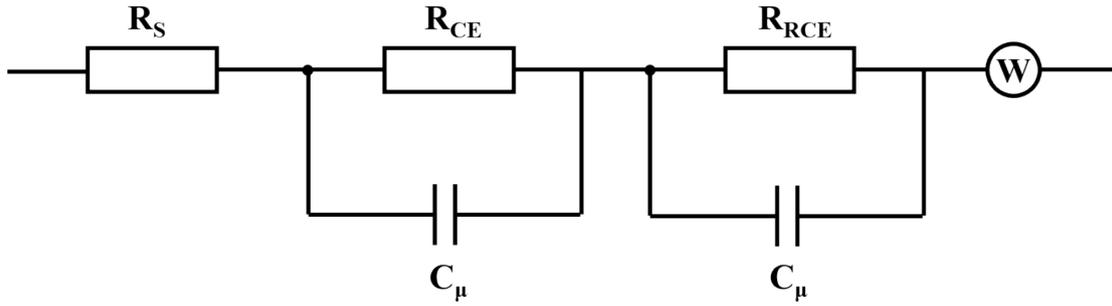
Figure S7. Equivalent circuit for EIS data fitting.
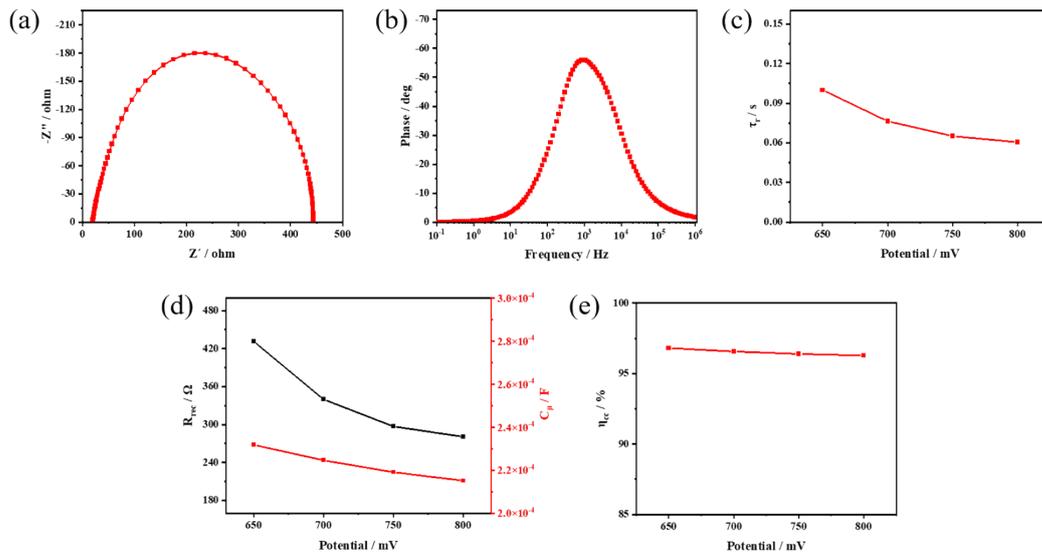
Figure S8. Nyquist plots (a) and bode plots (b) of DSSCs based on JM202. (d) interfacial charge-recombination resistance (Rrec), and chemical capacitance (Cμ), (e) electron lifetime (τ_r), (f) Charge-collection efficiency (η_{cc})fitted from impedance analysis under a series applied bias for DSSCs based on dye JM202.