Supplementary Information

Investigation of cobalt redox mediators and effects of TiO_2 film topology in dye-sensitized solar cells

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Figure S1. Steady-state absorption of ▼ Co^{II}(dMeObpy)₃, ▲ Co^{II}(dclbpy)₃, ◆ Co^{II}(dclphen)₃ and ,
Co^{II}(bpy)₃. A 0.1 cm pathlength cuvette and 15 mM solutions were used.



Figure S2. Cyclic voltammetry curves of \blacktriangle Co^{II/III}(dclbpy)₃, \blacktriangledown Co^{II/III}(dMeO-bpy)₃, \bullet Co^{II/III}(bpy)₃ and \bullet Co^{II/III}(dclphen)₃. Solutions with concentration of 15 mM Co^{II} were used and a solution of 15 mM ferrocene was used as reference.

Table S1. Current-voltage data of DSSCs containing working electrodes with different thicknesses. Transparent paste was 60 wt % Dyesol 18NR-T batch 300 + 36 wt % terpineol + 4 wt % ethylecellulose (Paste B). The transparent layer is denoted tr and scattering layer is denoted sc. The optimized concentration is marked in red. A 0.7 cm * 0.7 cm black mask is used.

	\mathbf{V}_{oc} / \mathbf{V}	J _{sc} / mAcm ⁻²	FF	η / %	thickness / µm
2 tr	0.865	10.40	0.676	6.10	6.8
2 tr+1 sc	0.835	11.30	0.685	6.48	10.2
3 tr+1 sc	0.825	10.56	0.646	5.63	12.7

Table S2. Current-voltage data of DSSCs containing working electrodes with different thicknesses. The transparent paste used for fabrication of the working electrodes was 40 wt % Dyesol 18NR-T batch 300 + 54 wt % terpineol + 6 wt % ethylcellulose (Paste C). A transparent layer is denoted tr and a scattering layer by sc. The optimized concentration is marked in red. A 7 cm * 7 cm black mask is used.

	V _{oc} / V	J _{sc} /	FF	η / %	Thickness
1 tr	0.865	6.49	0.625	3.51	3.04
1 tr +1 sc	0.915	9.44	0.605	5.23	5.01
2 tr+1 sc	0.890	10.38	0.663	6.13	7.64
3 tr+1 sc	0.885	9.36	0.635	5.26	9.55



Figure S3. Electron transport time as a function of the photo-current under short-circuit conditions for DSSCs sensitized with D35 employing different cobalt-based electrolytes. The traces symbolize \triangle Co^{II/III}(dclbpy)₃, \bigtriangledown Co^{II/III}(dMeO-bpy)₃, \blacklozenge Co^{II/III}(dclphen)₃, \bullet Co^{II/III}(bpy)₃ and \blacksquare Co^{II/III}(bpy)₃ electrolytes. A mixture of 40 wt% Ethylene carbonate + 60 wt% acetonitrile was used as solvent for the electrolyte. For \bullet Co^{II/III}(bpy)₃ acetonitrile was used as solvent.



Figure S4. Charge extraction as a function of the voltage of the DSSCs sensitized with D35 employing different cobalt-based electrolytes. The traces symbolize \blacktriangle Co^{II/III}(dclbpy)₃, \checkmark Co^{II/III}(dMeO-bpy)₃, \blacklozenge Co^{II/III}(dclphen)₃, \bullet Co^{II/III}(dpp)₃ and \blacksquare Co^{II/III}(bpy)₃ electrolytes. A mixture of 40 wt% Ethylene carbonate + 60 wt% acetonitrile was used as solvent for the electrolyte. For \bullet Co^{II/III}(bpy)₃ acetonitrile was used as solvent. Charge extraction experiments were performed to obtain the relation between charge and potential of the DSSC and could be estimated from the electron lifetime and transport time results. The differences in the presented curves in Figure S4 reflect the differences in the redox potentials and the obtained trend is almost the same as the differences in the redox potential values. The trap charge distribution, which was obtained from the charge-extraction experiments, is not affected by the differences in energy levels of the redox couples.

Table S3. Current-voltage characterization of DSSCs manufactured from the diluted pastes (B and C). All of the solar cells were prepared with working electrodes consisting of two transparent layers and one scattering layer. Two different paste concentrations lead to different thicknesses.

	Voc / V	Jsc / mAcm ⁻²	FF	η / %	thickness / µm
1	0,89	10,377	0,663	6,12	7.64
2	0,885	10,29	0,636	5,789	7.7
3	0,83	11,46	0,685	6,48	10.1
4	0,86	10,5	0,693	6,26	10.3
5	0,865	10,43	0,676	6,1	10
6	0,85	10,75	0,707	6,46	10.2
7	0,83	10,72	0,676	6,02	10.5
8	0,825	10,28	0,697	5,91	10.3
9	0,875	9,708	0,709	6,02	10.2
10	0,885	10,024	0,679	6,02	9.96
11	0,885	10,166	0,672	6,04	10.2
12	0,86	10,26	0,695	6,13	10.35
13	0,85	10,4	0,701	6,2	10.0
14	0,845	10,32	0,683	5,96	10.1
15	0,86	10,49	0,691	6,23	10.3
Average	0,86	10,41	0,684	6,12	
STDEV	0,022	0,38	0,019	0,19	



Figure S5. SEM micrographs of a TiO_2 electrode screen printed with Paste A at different magnifications and profilometer data (bottom right) showing very few cracks.