Selective co-sensitization approach to increase photon conversion efficiency and electron lifetime in dye-sensitized solar cells

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Experimental

Device fabrication: The C106 (ORGANICA Feinchemie GmbH Wolfen), D131 (gifted by Mitsubishi Paper Mills Limited) and C106+D131 co-sensitized devices were fabricated as reported elsewhere.\(^1\) Briefly, the FTO glass (2.2 mm thickness, 14 Ω/sq, Pilkington) used as current collector was first cleaned, then immersed into a 40 mM aqueous TiCl\(_4\) solution at 70 °C for 30 min and rinsed with water and ethanol. 10μm-thick transparent TiO\(_2\) layers (Dyesol 18NR-T, average nanoparticle size: 20 nm) were screen-printed on the FTO glass plates (mesh count is 90T mesh/cm, active areas are 0.16 cm\(^2\)). After drying at 125 °C, 5μm-thick scattering TiO\(_2\) layers (Dyesol WER2-O paste, average nanoparticle size: 150-250 nm) were superimposed by screen-printing. The electrodes coated with the TiO\(_2\) pastes were gradually annealed under air at 125 °C for 10 min, 325 °C for 5 min, at 375 °C for 5 min, and at 450 °C for 15 min, and finally, at 500 °C for 15 min. The TiO\(_2\) films which were again treated with 40 mM TiCl\(_4\) solution for 30 min at 70 °C, were rinsed with DI water and ethanol and annealed at 500 °C for 30 min. After cooling to room temperature, the TiO\(_2\) photoanodes of optimum devices were made by immersing into: a) Solution A - 0.3 mM C106 and 0.3 mM chenodeoxycholic acid in a mixture of dimethylformamide (DMF), acetonitrile (ACN) and tert-butyl alcohol (TBA) (volume ratio, 0.2:1:1) for 12 h; b) Solution B - 0.2 mM D131 dye solution in a mixture of ACN and TBA (volume ratio, 1:1) for 4 h and c) Solution A for 12 h and then solution B for 4 h for co-sensitization. The dye sensitized TiO\(_2\) photoanode and the Pt-coated FTO glass counter electrode were sandwiched together using a 25 μm-thick
transparent Surlyn film (Meltonix 1170-25, Solaronix). The electrolyte was injected through the hole at the back of the counter electrode via vacuum backfilling. The electrolyte employed was a solution of containing 1.0 mM 1,3-dimethylimidazolium iodide (DMII), 50 mM LiI, 30 mM I₂, 0.5 mM tert-butylpyridine, and 0.1 mM guanidinium thiocyanate (GNCS) in the mixed solvent of acetonitrile and valeronitrile (v/v, 85/15). ² Finally, the hole was sealed using 25 μm-thick surlyn and a cover glass (0.1 mm thickness) to avoid leakage of the electrolyte.

**Photovoltaic measurements:** Photocurrent density - photovoltage (J-V) characteristics were measured under an illumination of AM 1.5 (100 mW cm⁻²) using a solar simulator (San-EI Electric, XEC-301S) equipped with a 450 W xenon lamp which was coupled with an Agilent semiconductor parameter analyzer (4155C). The power of the simulated light was calibrated to100 mW/cm² by using a silicon reference cell (Fraunhofer) and monitored using a power meter throughout the testing. A black mask (6 mm x 6 mm) was used in the subsequent photovoltaic studies to avoid the effects of diffusive light on cell performance. The reported values are calculated based on the average of three batches of devices with identical composition and fabrication procedure. An IPCE measurement system equipped with a xenon lamp (Oriel 66902, 300 W), a monochromator (Newport 66902). Incident light intensity was calibrated using a photodiode detector (silicon calibrated detector, Newport). All the measurements were performed in air. UV-Vis absorption spectra were collected with a Shimadzu UV3600 spectrophotometer. The photoelectrochemical impedance spectroscopy and intensity modulated photovoltage spectroscopy (IMVS) for DSSCs were carried out with an Autolab potentiostat (PGSTAT
302) using together with a white LED (LUXEON). For IMVS, the modulation of the LED light is set 10% of the DC light intensity.
**Figure S1.** Reproducibility of the PV characteristics recorded on co-sensitized C106 + D131 devices fabricated under identical conditions.

![Graph showing PV characteristics](image-url)
Figure S2. Reproducibility of the PV characteristics recorded on C106 sensitized devices fabricated under identical conditions.
Figure S3. Reproducibility of the PV characteristics recorded on D131 sensitized devices fabricated under identical conditions.
Figure S4. Nyquist plot of: a) C106, b) D131 & c) C106+D131 sensitized devices.
**Figure S5.** Electron lifetime obtained from photoelectrochemical impedance spectroscopy.

![Electron lifetime vs Voc](image)

**References**
