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Electronic Supporting Information for:

The Impact of Non-Uniform Photogeneration on Mass Transport in Dye-Sensitised Solar Cells

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Fig. S1 a) Transmission spectrum of colour filters used in study: blue filter (top spectrum, C3C8, 2), green filter (middle spectrum, 3C8), red filter (bottom spectrum, KC13, 2). b) Relative colour spectrum of WLED (warm white, Bridgelux BXRC-27G10K0-L-03) used in this study. Figure obtained from manufacturer's product data sheet.



Fig. S2 Comparison of τ_{tr} electron transport values (Fig. 6) versus t_{COL} collection limited photocurrent turn-on transient (Fig. 6). Measurements under blue EE-illumination.



Fig. S3 Comparison of t_{COL} versus light intensity.



Fig. S4 Comparison of J_{COL} versus J_{SS} .



Fig. S5 Excess charge density extracted from short-circuit (Q_{SC}), measured under SE-blue illumination, for 6.5 µm prepared with compact TiO₂ blocking layer.



Fig. S6 Current decay time constant (τ_{DJ}) as a function of J_{SS} . τ_{DJ} is equivalent to τ_{tr} for EEillumination, where the current decay constant strongly fitted to a mono-exponential function. Some transients required fitting to a biexponential function, for which the initial decay time constant is presented.



Fig. S7 Normalised small perturbation photocurrent transient at 2 Suns steady state illumination and short-circuit. Transients normalised at J_{SS} for comparison. Comparing various FTO-blocking layers: compact TiO₂ blocking layer (BL), TiCl₄ pre-treatment (TICl₄) and untreated FTO (FTO).



Fig. S8 Comparison of relative spectral irradiance distribution for Newport AM1.5 solar simulator and WLED with various bandpass filters. Units are given in absolute irradiance. 5 sets of repeats overlayed. Measured using UV-Vis spectrometer fitted with integrating sphere. The TEC8 glass used for CE has lower transmittance than TEC15. Also, CE transmittance will vary with uniformity of back electrode sealing and uniformity of Pt layer.



Fig. S9 Photocurrent turn-on transients comparing high efficiency (>6.5% PCE under 1 Sun AM1.5) LEG4 cobalt DSSC with ~5µm TiO2 (DSL-18NRT) film. This shows similar equivalent trends to Fig.4 in the main paper.



Fig. S10 Photocurrent turn-on transients comparing efficient (>6.5% PCE under 1 Sun AM1.5) LEG4-devices with 5μm TiO2 (DSL-30NRD) + 2μm TiO2 scattering layer (WER2-O) film. Comparison effect of 40mM and 26.7mM TiCl4 concentration used in post-treatment of TiO2 films, displayed on left and right, respectively. Indistinguishable JV performance at 1 Sun. Slightly noticeable improvement in mass transport at 2 Suns. Devices mainly still limited by light absorption. This shows similar equivalent trends to Fig.4 in the main paper.



Fig. S11 Charge extraction current decay transients comparing high efficiency (>6.5% PCE at 1 Sun AM1.5) LEG4 cobalt DSSC. Normalised curve shown on right. This shows similar equivalent trends to Fig.4 in the main paper.



Fig. S12 J-Vs of performance optimised LEG4 DSSCs. All devices had ~5.6um±0.2um TiO2 films. NRT1 fabricated using DSL18NRT paste. 4T2 and 2T2 fabricated using DSL30NRD paste and WER2-O scattering layer (5.6+2um). TiCl4 post-treatment concentration varied for 4T2 and 2T2 as 40mM and 26.7mM; little difference in JV performance at 1 Sun. Measured under 1 Sun AM1.5 solar simulator (Newport).



Fig. S13 J-Vs comparing high efficiency LEG4 device, with similar components including 18NRT TiO2 based films. Measured under WLED with band-pass filters, under 2 Suns intensity. SE/EE illumination and illumination wavelength compared. J-V hysteresis increases with stronger SE-abs.



Fig. S14 Small perturbation photocurrent transients for 6.5% LEG4-cobalt NRT-based DSSC. This shows similar equivalent trends to Fig.10 in the main paper.



Fig. S15 Small perturbation photocurrent transients for 6.6% LEG4-cobalt 30NRD+WER3-Obased DSSC. This shows similar equivalent trends to Fig.10 in the main paper.