

## Supporting Information

### Wavelength Conversion Effect-Assisted Dye-Sensitized Solar Cells for Enhanced Solar Light Harvesting

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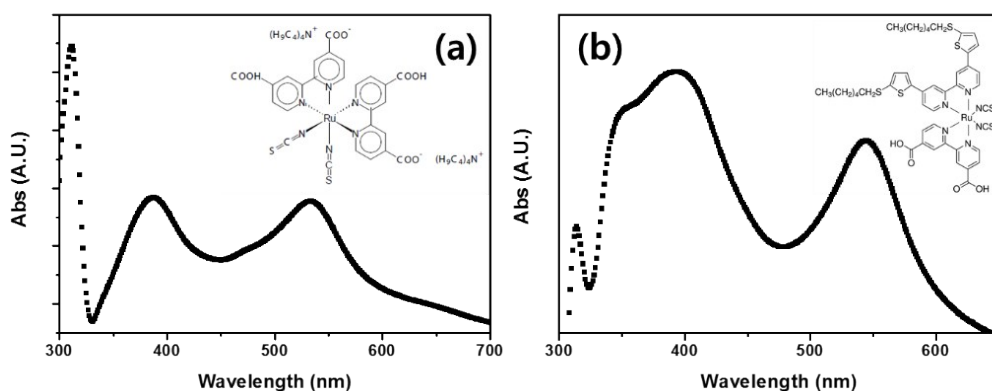


Figure S1. Chemical structure and UV-vis spectrum of (A) N719 dye and (b) C106 dye.

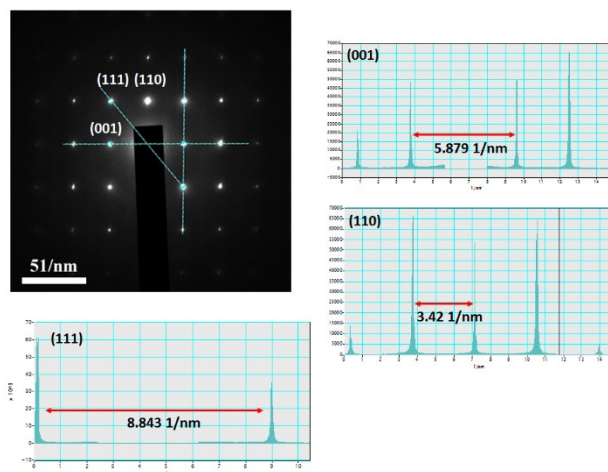


Figure S2. Lattice distance calculations for the  $\beta$ - $\text{NaYF}_4:\text{Yb}^{3+},\text{Er}^{3+}$  phosphor microcrystals.

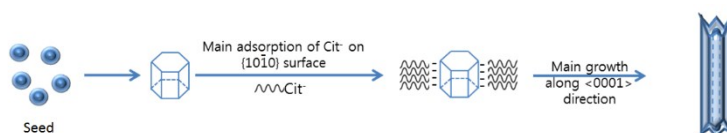


Figure S3. Crystal growth mechanism of the  $\beta$ - $\text{NaYF}_4:\text{Yb}^{3+},\text{Er}^{3+}$  phosphor microcrystals.

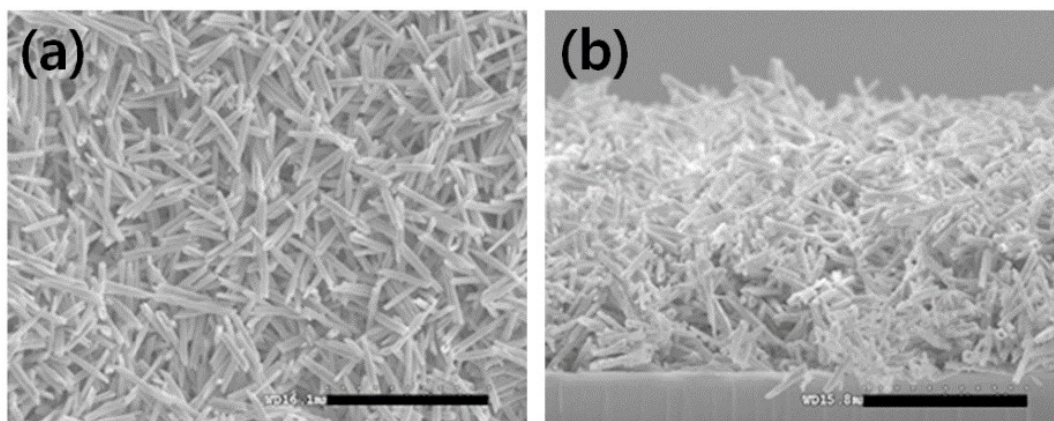


Figure S4. Top and cross-sectional SEM images of the  $\beta$ - $\text{NaYF}_4:\text{Yb}^{3+},\text{Er}^{3+}$  phosphor microcrystals used as back-reflection films, on glass substrates (scale bar:  $20\ \mu\text{m}$ ).

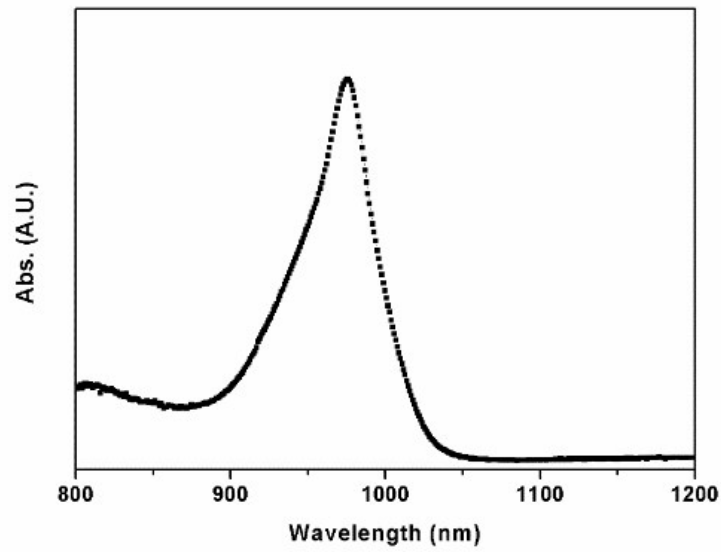


Figure S5. UV-Vis spectrum of the  $\beta$ - $\text{NaYF}_4:\text{Yb}^{3+},\text{Er}^{3+}$  phosphor microcrystals.

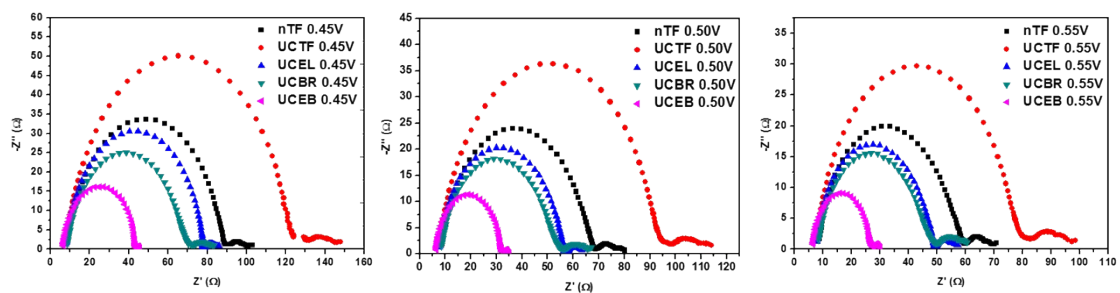


Figure S6. Comparison of Nyquist plots for various kinds of samples under IR range ( $> 815 \text{ nm}$ ).

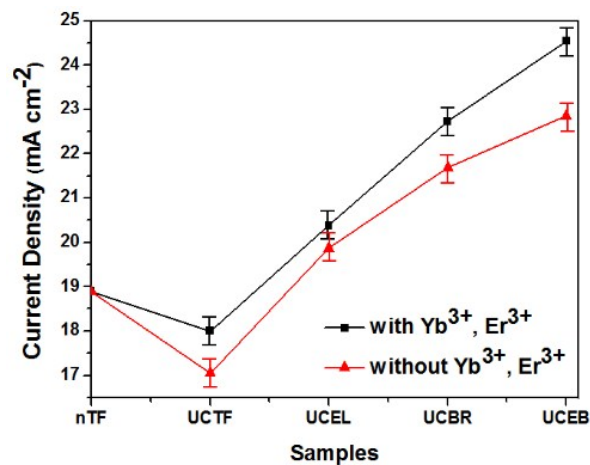


Figure S7. Comparison of the photocurrent density for various kinds of samples with and without  $\text{Yb}^{3+},\text{Er}^{3+}$ .

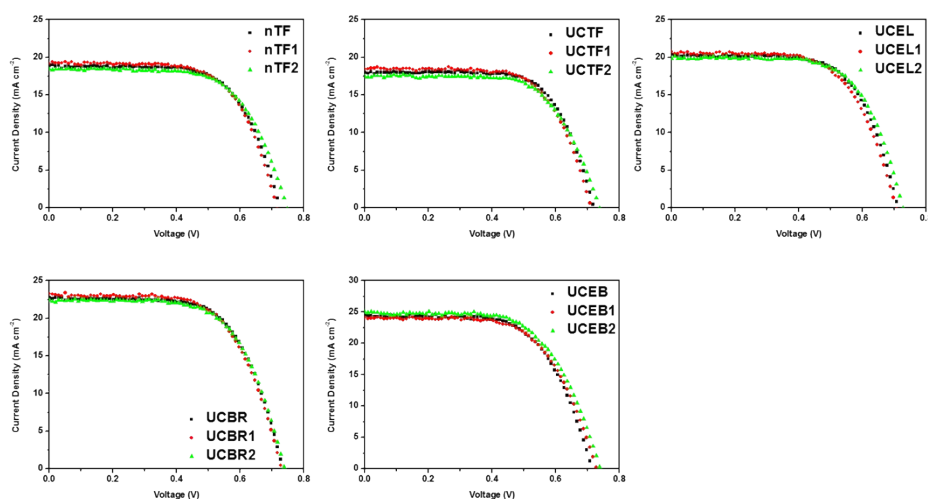


Figure S8. Comparison of I-V curves for samples' reproducibility under 1-sun illumination. Each samples was measured with over 3 times for reproducibility.

## MATERIALS AND CHARACTERIZATIONS

**Materials:** Yttrium (III) nitrate tetrahydrate ( $Y(NO_3)_3 \cdot 4H_2O$ , 99.99%, Aldrich), ytterbium (III) nitrate pentahydrate ( $Yb(NO_3)_3 \cdot 5H_2O$ , 99.9%, Aldrich), erbium (III) nitrate pentahydrate ( $Er(NO_3)_3 \cdot 5H_2O$ , 99.99%, Aesar), and sodium fluoride ( $NaF$ , > 98.5%, Ajax) were used as  $Y^{3+}$ ,  $Yb^{3+}$ ,  $Er^{3+}$ , and  $F^-$  sources, respectively. Sodium citrate ( $Na_3C_6H_5O_7 \cdot 2H_2O$ , > 99.0%, Aldrich) was used as a chelating agent, whereas nitric acid ( $HNO_3$ , 60.0-62.0%, Daejung) was used to control pH. Branched polyethylenimine (PEI,  $C_2H_5N$ , typical Mw 25,000, Aldrich), was used for promoting adhesion between the prepared  $\beta$ - $NaYF_4:Yb^{3+}, Er^{3+}$  phosphor microcrystal powders and the glass substrate. All the chemical reagents were used as-received without further purification. Ultrahigh purity deionized water (> 18 MW, Millipore) was used for all the experiments.

**Characterization and Photovoltaic Performance Measurements of DSSCs:** A HRTEM (JEM 2100F, Japan) equipped with an EDS detector, operated at an accelerating voltage of 200 kV was used for analyzing the crystal structure and SAED patterns of the samples. In the HRTEM

experiments, the electron beam was incident in a direction perpendicular to the 1D microstructure. The  $\beta$ -NaYF<sub>4</sub>:Yb<sup>3+</sup>,Er<sup>3+</sup> phosphor microcrystals were transferred onto TEM copper grids by directly dropping solutions of the  $\beta$ -NaYF<sub>4</sub>:Yb<sup>3+</sup>,Er<sup>3+</sup> phosphor microcrystals dispersed in ethanol solution. The morphology of the products was examined with a field emission scanning electron microscope (FESEM, Hitachi S-4300, Japan) operated at an acceleration voltage of 20 kV. A platinum/palladium alloy (w/w, 8:2) with a thickness of about 15 nm was deposited over the samples. The crystal structure of the synthesized materials was characterized by XRD (Rigaku miniFlex-2 desktop X-Ray diffractometer, Cu K $\alpha$  radiation with  $\lambda = 0.154056$  nm, Japan) operating at 30 kV and 15 mA, with a scanning rate of 0.02° per step in the  $2\theta$  range of  $10^\circ \leq 2\theta \leq 80^\circ$ .

The PL spectra and images of single phosphors as well as single particles were acquired using a homemade microscope setup, composed of an inverted microscope (TE2000-U, Nikon), an NIR (980 nm) diode laser (P161-600-980A, EM4), and two cameras, including a charge coupled device (CCD) camera (PIXIS 400BR, Princeton Instruments) attached to a monochromator (HoloSpec f/1.8i, Kaiser Optical Systems) and an electron multiplying charge coupled device (EMCCD) camera (DV897DCS-BV, iXon, Andor Technology), for acquiring luminescence spectra and for imaging, respectively. The focused 980-nm laser beam ( $\sim 100$  W cm<sup>-2</sup>), which passes through the microscope objective (Plan Fluor, 20x, NA 0.50, Nikon), excited the products on the coverslip. The luminescence exhibited by the excited products was collected by an optical fiber, and finally detected by the CCD camera. In addition, single phosphors were imaged by EMCCD through another pass way without the optical fiber.

I-V performance of DSSCs was measured under 1-sun conditions (AM 1.5 G, 100 mW cm<sup>-2</sup>) using a solar simulator (Newport, 91160-1000) equipped with a 300 W Xenon lamp and a

Keithley (Model 2400) source meter. I-V performance under NIR range was measured using high performance long-pass filter (66-235, Edmond), of which transmission wavelength is from 815 to 1650 nm. All photovoltaic performances were determined after calibrating with a silicon reference cell. The solar light was incident on a 0.2 cm × 0.8 cm area of the photoanode surface. The other areas of the surface were masked to avoid overestimation of the I-V performances. All the average values were obtained from measurements in triplicate. The IPCE and electrochemical impedance spectroscopy (EIS) data for the DSSCs were recorded under the same light conditions as the I-V performance measurements. The EIS measurements were conducted under 1-sun illuminated condition in the 1 MHz–200 mHz frequency range, by applying at – 0.70 V AC voltage using a DIGITAL-AVR potentiostat (UPS BANK, Korea) operated in a two-electrode mode. The IPCE measurements were carried out using QEX7 (PV Measurements, Inc.) with a chopping speed of 10 Hz.