

Electronic Supplementary Information

Broadband and ultrahigh optical haze thin films with self-aggregated alumina nanowire bundles for photovoltaic applications

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S1. Influence of etching time on the morphologies of structure.

We observed the morphologies of anodized aluminum oxide (AAO) template as a function of etching time. As shown in the SEM images (Fig. S1), the alumina pore walls start to break down at 42min-etching time. However, due to the insufficient etching of alumina pore wall, AAO structure did not entirely break into isolated nanowires nor collapsed. The non-collapsed alumina structures (or alumina nanopore arrays) did not show diffusive transmittance because the subwavelength periodic structure ($p = \sim 100\text{nm}$) can be regarded as a homogeneous effective medium. Therefore, a minimum etching time of 43min is necessary to obtain the hazy film consists of collapsed and aggregated alumina nanowire structure.

Fig. S1 SEM images of etching time-controlled AAO (anodized aluminum oxide) templates.

S2. FDTD simulation details

We performed full-wave three-dimensional FDTD simulations, employing a commercial software package (Lumerical). Broadband plane wave source (300~800nm) was launched toward an inverted type organic photovoltaic (OPV) device which is composed of ITO (160nm)/PFN (5 nm)/PTB7:PC₇₀BM (90 nm)/MoO₃ (8 nm)/Ag (100 nm) layers. A plane power monitor located above the source acquires reflectance from the device. We used perfectly matched layers (PML) boundary condition in the vertical direction and periodic boundary condition in the horizontal directions. We set a background index of 1.5, corresponding to typical glass substrate.

If there is no haze film on a glass surface of OPV device, the incident light does not undergo scattering at air/glass interface. Therefore, we launched a normal incident plane wave from the top to calculate the reflectance of bare OPV device. On the other hand, the incident light was significantly scattered and divided into numerous light rays when the haze film was attached on the glass surface. To reflect this scattering effect of the haze film in simulation, measured angular dependent light scattering intensities were utilized as intensities of light rays at different angles. Because direct sunlight and light sources used in our experiments were unpolarized, we calculated and simply averaged the reflectance for transverse electric (TE) and transverse magnetic (TM) polarized incident waves.

Fig. S2 Simulated reflectance spectra of the PTB7:PC₇₀BM-based organic photovoltaic device with and without haze film for (a) TM and (b) TE polarized light.

S3. Measured J_{sc} from solar simulator vs calculated J_{sc} from EQE spectra

Based on the equation $J_{sc} = \int qF(\lambda)EQE(\lambda)d\lambda$, where q is the electron charge, we could extract the J_{sc} from the EQE spectra by integrating the product of the incident photon flux density $F(\lambda)$ and $EQE(\lambda)$ of the cell over the wavelength (λ) of the incident light. Table S1 compares the J_{sc} experimentally obtained from the J - V characteristics vs J_{sc} calculated from the spectral EQE spectra. The *exp.* J_{sc} and *cal.* J_{sc} were highly similar within measurement error possibly occurring in ambient measurement conditions.

Table. S1 Comparison of experimentally obtained J_{sc} (from J-V characteristics) vs calculated J_{sc} (from spectral EQE spectra).

	J_{sc} (mAcm ⁻²)	J_{sc} (EQE) (mAcm ⁻²)	J_{sc} (EQE)/ J_{sc} (J-V)	ΔJ_{sc} (%)	ΔJ_{sc} (EQE) (%)
Bare	16.19	15.23	0.941	-	-
Haze A	18.10	16.87	0.931	+11.79%	+10.74%
Haze B	17.69	16.66	0.942	+9.27%	+9.35%
Haze C	16.98	15.94	0.939	+4.88%	+4.65%