Supplementary Information

Removal of Polymeric Top Skins

Polymeric top skins are noted in both dip and spray LbL porous films, and few open pores exist on the surface (see **Figure S1(a)** and **S1(b)**). To obtain templates with a pore-rich top surface, erosion treatments are conducted to create an open porous morphology (see **Figure S1(c)** and **S1(d)**).

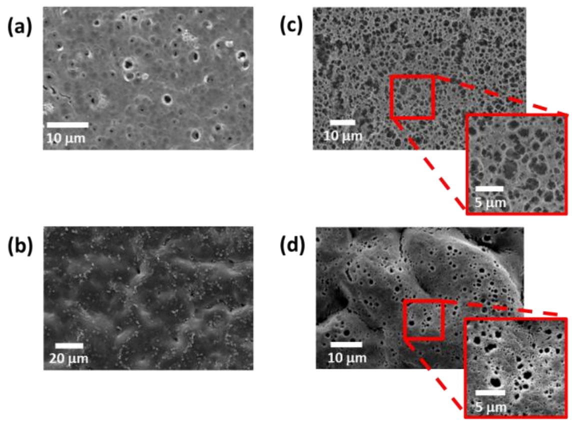

Figure S1. SEM images of dip (a, c) and spray (b, d) $(LPEI/PAA)_n$ templates before top crust removal (a, b) and after top crust removal (c, d).

Optimization of the fabrication processes for dip and spray LbL-templated photoanodes

To achieve 10-15 µm thick and high dye loading photoanodes, two sets of dip $(LPEI/PAA)_{40}$ and one set of spray $(LPEI/PAA)_{110}$ are deposited to remain high dye loading (**Figure 2(a)(b)** and **Figure S2**). Also, two sets of dip $(LPEI/PAA/LPEI/phage)_{26}$ and one set of spray $(LPEI/PAA/LPEI/phage)_{85}$ are deposited (**Figure S3**).

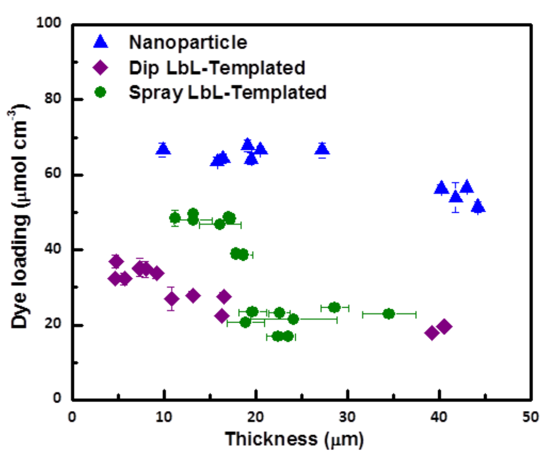

Figure S2. Dye loading of nanoparticle, dip and spray LbL-templated photoanodes as a function of film thickness.

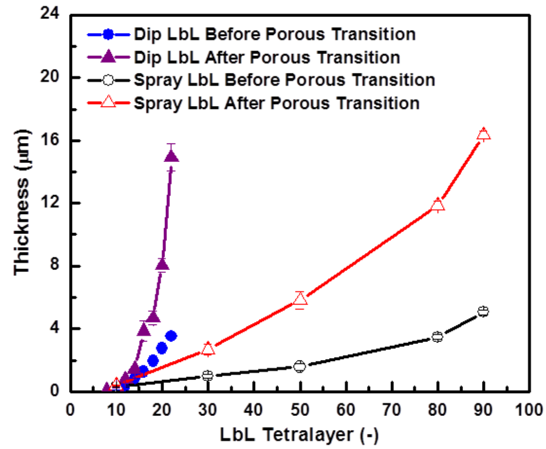

Figure S3. The growth curves of dip and spray $(LPEI/PAA/LPEI/phage)_m$ deposition before and after porous transition.

Calculation of the volume percentage of phage in LbL films

A small aliquot (100 µL) of the phage dip bath was removed after every 5 tetralayers of film deposition in order to measure the concentration of the phage in the dip bath. Concentration measurements, as determined by UV-Vis spectroscopy, indicated that the phage concentration was depleted by about ~10%. An aliquot of the phage rinse bath was also removed to determine the concentration of phage lost but not deposited on the film. When combined with information about how much substrate area was exposed to the dipping baths and the film thickness of the dipped film on the substrate, the number of phage per film volume could be calculated. A volume fraction was calculated from this density by approximating a single phage as a cylinder 6 nm in diameter and 860 nm in length.

Optimization the spray time of the phage deposition

By using different OG-labeled phage spraying times on $(LPEI/PAA)_{4.5}$ film, different florescent signal intensities of OG-labeled phage in the center of films are observed in **Figure S4**. A five-second spray time is chosen to deposit the phage to get high loading without wasting material.

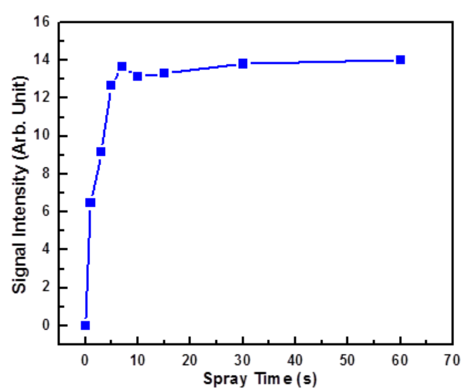

Figure S4. Signal intensities of spraying different time of OG-labeled phage on $(LPEI/PAA)_{4.5}$.

Calculation of Electron Diffusion Length - Transmission Line Model

Transmission line model is shown in **Figure S5**. A simplified version was used for fitting the impedance data when certain parameters (such as those of the photoanode-FTO or electrolyte-FTO interfaces or Warburg diffusion) were not evident in the impedance spectra.

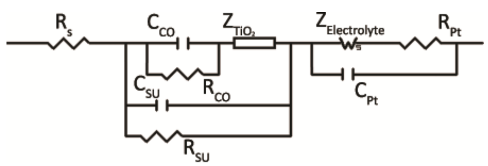

Figure S5. Equivalent circuit impedance model of DSSCs. R_s: Ohmic series resistance of the cell. R_{CO} and C_{CO}: Contact resistance and capacitance at the interface between the conducting substrate and the TiO_2 photoanode film. R_{SU} and C_{SU}: Charge transfer resistance and double layer capacitance at the substrate/electrolyte interface. R_{Pt} and C_{Pt}: Charge transfer resistance and double layer capacitance at the counter electrode-electrolyte interface. Z_{TiO2}: transmission line impedance of the TiO_2 photoanode film consisting of the elements R_T (resistivity of electron transport in the photoanode film), R_{REC} (charge recombination resistance at the TiO_2/dye/electrolyte interface), and C_μ (chemical capacitance of the photoanode film). $Z_{Electrolyte}$: mass transport impedance at the counter electrode.

Table S1. Additional photovoltaic data for DSSCs employing LbL-templated photoanodes to demonstrate the reproducibility.

Photoanodes Type	V_{OC} (mV)	FF (%)	J_{SC} (mA cm^{-2})	PCE (%)
Dip LbL	708	63.0	11.60	5.17
Spray LbL	712	58.2	13.40	5.55
Phage-containing dip LbL	727	59.6	14.19	6.15
Phage-containing spray LbL	675	63.2	14.24	6.06

Figure S6. The fabricated device with the black mask we measured in the article.