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Electronic Supplementary Information for

Edge-selectively antimony-doped graphene nanoplatelets as an outstanding counter electrode with an unusual electrochemical stability for dye-sensitized solar cells employing cobalt electrolyte

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Preparation of SbGnPs electrodes. Homogeneously dispersed 0.1 wt.% sample powders in 2-propanol solution were obtained by ultrasonication for 30 min. The resultant solution was deposited directly onto FTO/glass (TEC-8, Pilkington) using an e-spray technique. First, the sample dispersion solutions were loaded into a plastic syringe equipped with a 30-gauge stainless steel hypodermic needle. The needle was connected to a high voltage power supply (ESN-HV30). A voltage of ~ 4.5 kV was applied between a metal orifice and the conducting substrate at a distance of 9 cm. The feed rate was controlled by a syringe pump (KD Scientific Model 220) at a constant flow rate of 50 μL min^{-1}. The electric field overcame the surface tension of the droplets, resulting in minimization of numerous charged mists. The sample electrodes were sintered at 300 ^{o}C for 30 min at atmosphere prior to device fabrication. For the reference, Pt-FTO electrode was also prepared by deposition of *ca.* 30 μL cm^{-2} of H_2PtCl_6 solution (2 mg of H_2PtCl_6 in 1 mL of ethanol) and sintered at 400 °C for 15 min.

Fabrication of a symmetrical dummy cell. Symmetrical sandwich dummy cells were fabricated with two identical SbGnPs- and Pt-FTO sheets, which were separated by 25-μm thick Surlyn (Solaronix, Switzerland) tape as a sealant and spacer, leaving an active area of 0.6 × 0.6 cm^2. The sheet edges were coated by an ultrasonic soldering system (USS-9200, MBR Electronics) to improve the electrical contacts. The electrolyte solution was prepared with 0.22 M $Co(bpy)_3(BCN_4)_2$, 0.05 M $Co(bpy)_3(BCN_4)_3$, 0.1 M $LiClO_4$, and 0.8 M 4-*tert*-butylpyridine in acetonitrile. Co-complexes were prepared by a reported procedure.[S1] Electrolyte solution was introduced through a drilled hole on the CEs via vacuum backfilling. The hole was sealed with cover glass using a Surlyn seal.

Electrochemical analysis. All electrochemical measurements for electrocatalytic evaluation of materials were carried out with VersaSTAT 3 (Version 1.31), AMETEK, connected to a potentiostat under dark conditions at room temperature. EIS spectra were acquired in the frequency range from 10^6 to 0.1 Hz at an open circuit voltage of 0 V and AC modulation amplitude of 10 mV. The EIS data analysis was processed using Zplot/Zview2 software.

Fabrication and characterization of DSSCs. FTO plates were cleaned in detergent solution, water, and ethanol using an ultrasonic bath. FTO substrates were immersed in 40 mM aqueous $TiCl_4$ solution at 70 °C for 30 min and washed with water and ethanol. TiO_2 colloidal paste (Dyesol, 18NR-T) was screen-printed onto FTO/glass and sintered at 500 °C for 30 min in air. The thickness of the transparent layer was measured by using an Alpha-step 250 surface profilometer (Tencor Instruments, San Jose, CA), and a paste for the scattering layer containing 500-nm-sized anatase particles (ENB Korea, STP-500N) was deposited by doctor blade printing and then dried for 2 h at 25 °C. TiO_2 electrodes were sintered at 500 °C for 30 min. The resulting TiO_2 photoanode was immersed in THF/ethanol (v/v, 1/3) solution containing 0.2 mM of SGT-021 sensitizer based on a D-π-A structured zinc(II)-porphyrin and 0.6 mM of HC-A4 coadsorbent[S2] and then kept at room temperature for 2 h. The dye adsorbed TiO_2 photoanodes were assembled with SbGnP or Pt CEs using a thermal adhesive film (25 μm thick Surlyn, Du-Pont) as a spacer to produce a sandwich-type cell. Electrolyte solution was introduced through a drilled hole on the CEs via vacuum backfilling. The hole was sealed with cover glass using Surlyn. The light intensity was adjusted with a Si solar cell that was doubled-checked with an NREL-calibrated Si solar cell (PV Measurement Inc.). The applied potential and measured cell current were measured using a Keithley model 2400 digital source meter. Photoelectrochemical data were measured using a 1000-W xenon light source (Oriel, 91193) that was focused to give 100 mW cm^{-2} (1 sun at AM 1.5 G). The *J-V* characteristics of the cell under these conditions were determined by biasing the cell externally and measuring the generated photocurrent. This process was fully automated using Wavemetrics software. The measurement settling time between applying a voltage and measuring a current for the *J-V* characterization of DSSCs was fixed to 80 ms.

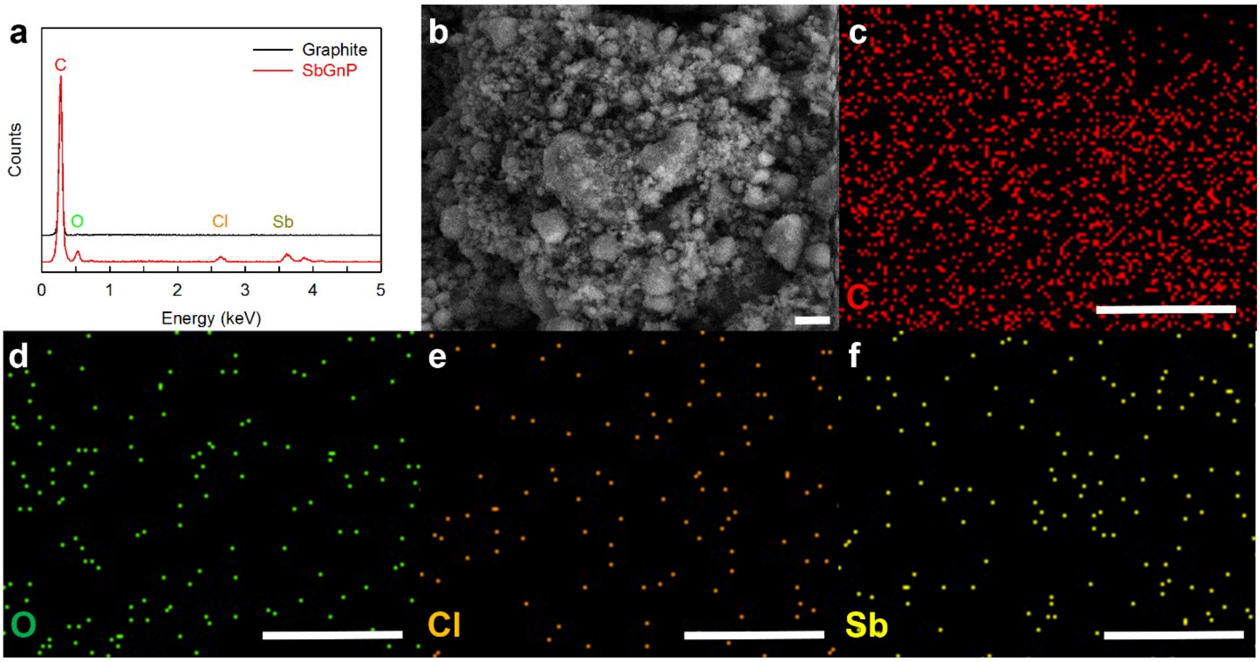


Figure S1. (a) Energy dispersive x-ray (EDX) spectra of the pristine graphite and SbGnPs. (b) FE-SEM image of SbGnPs. Corresponding element mappings: (c) carbon; (d) oxygen; (e) chloride; (f) antimony. Scale bars are 500 nm.

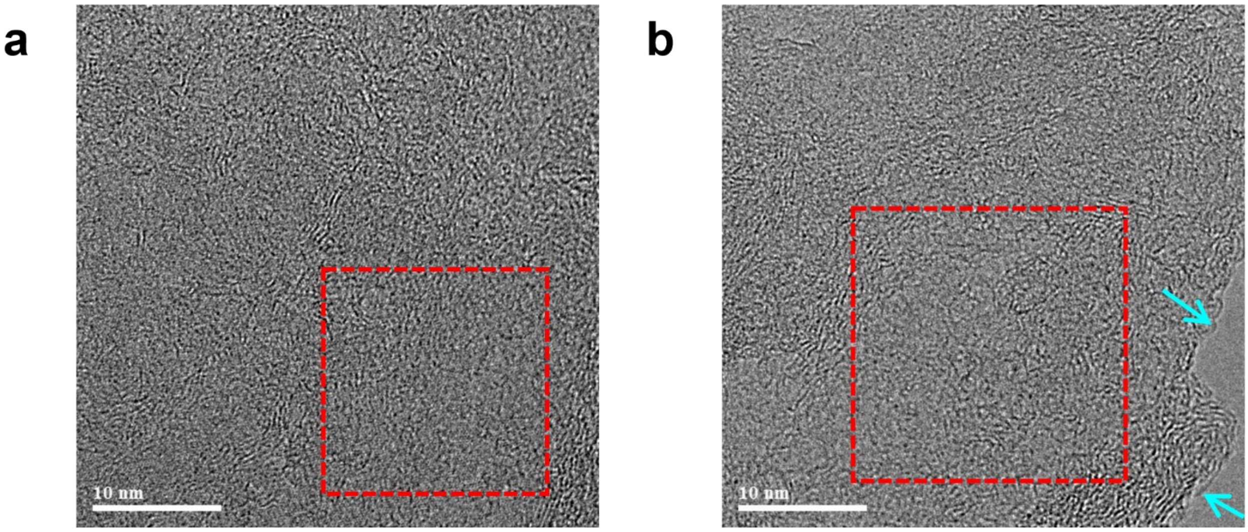
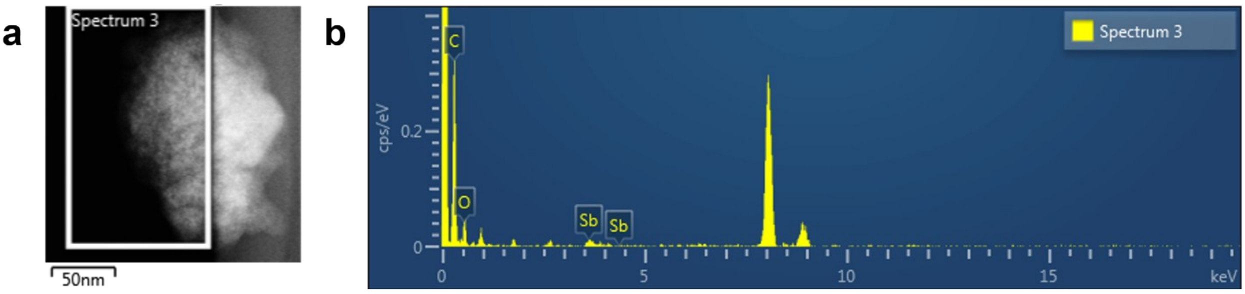


Figure S2. HR-TEM images of SbGnPs.



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Element	Line Type	k factor	Absorption Correction	Wt.%	Wt.% Sigma
C	K series	2.75165	1.00	87.16	2.45
O	K series	2.00744	1.00	8.90	1.89
Sb	L series	1.93957	1.00	3.94	1.74
Total:				100.00	

Figure S3. (a) Dark-field (DF) TEM image of SbGnPs. Scale bars are 50 nm. (b) Energy-dispersive X-ray (EDX) spectra of SbGnPs. (c) The table shows corresponding element contents (wt.%).

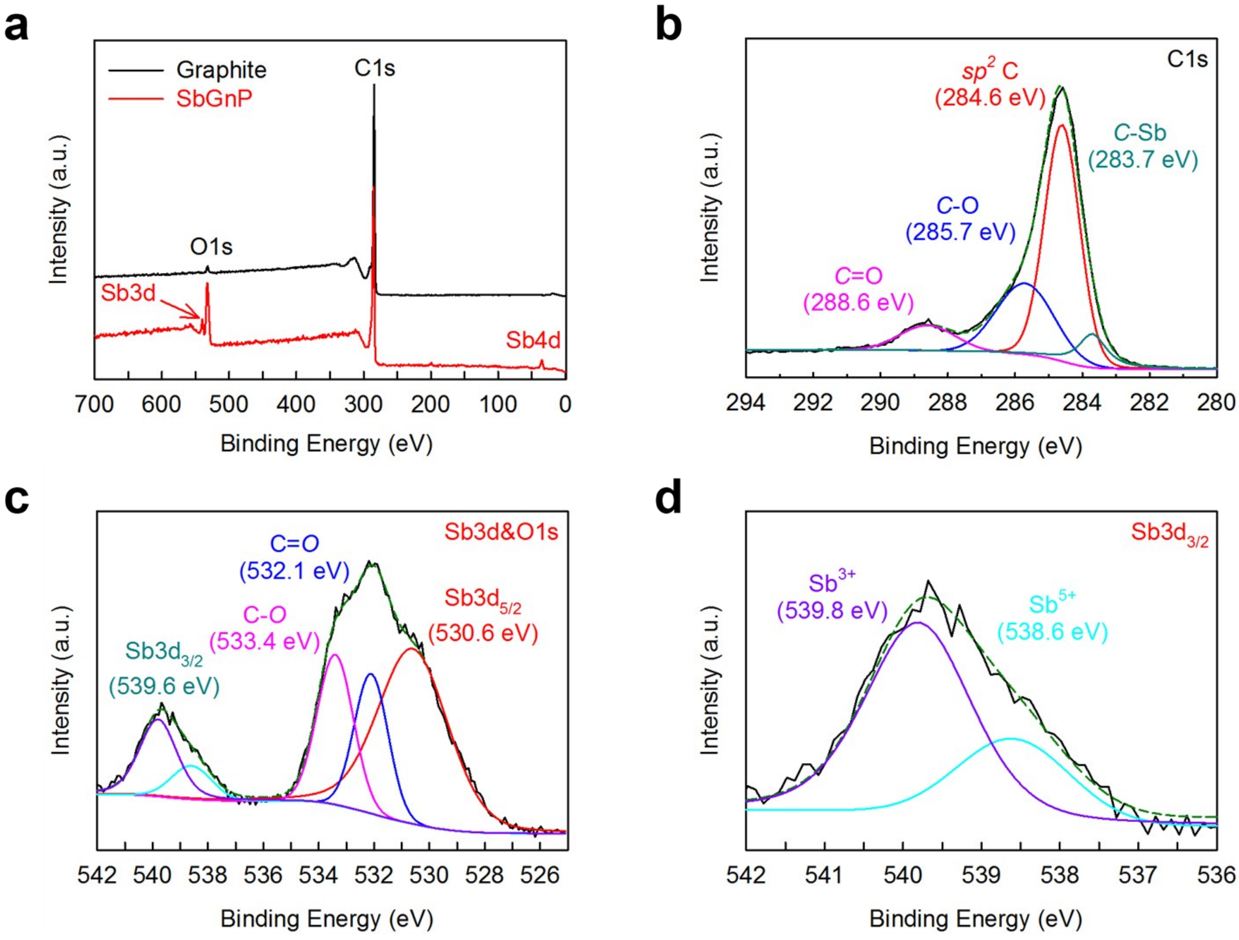

Figure S4. XPS spectra: (a) Survey of the pristine graphite and SbGnPs; (b) C1s; (c) Sb3d and O1s; (d) Sb3d$_{3/2}$.

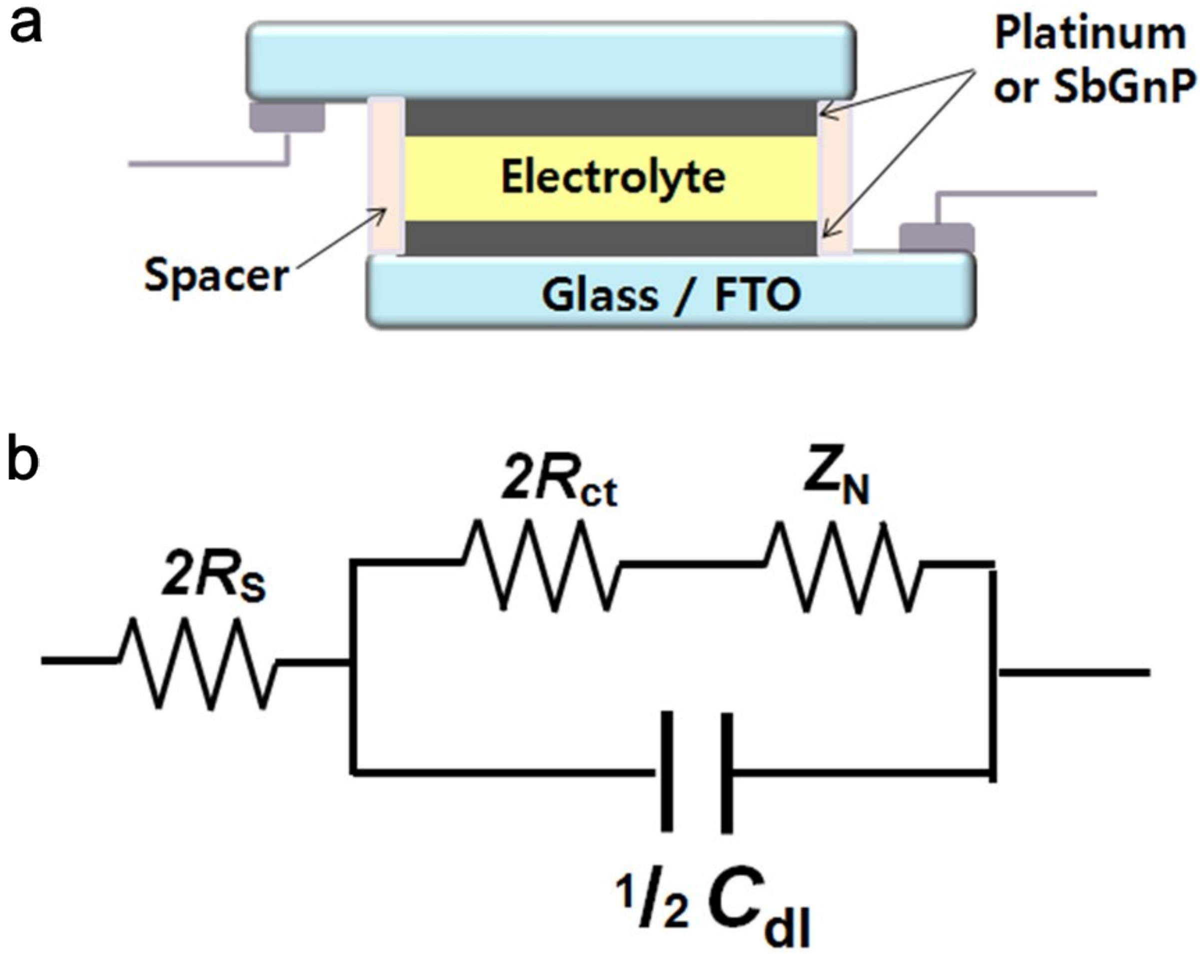

Figure S5. (a) A symmetrical dummy cell fabricated with two identical electrodes. (b) An equivalent circuit (EC) for fitting the impedance spectra having two characteristic semicircles.[S3]

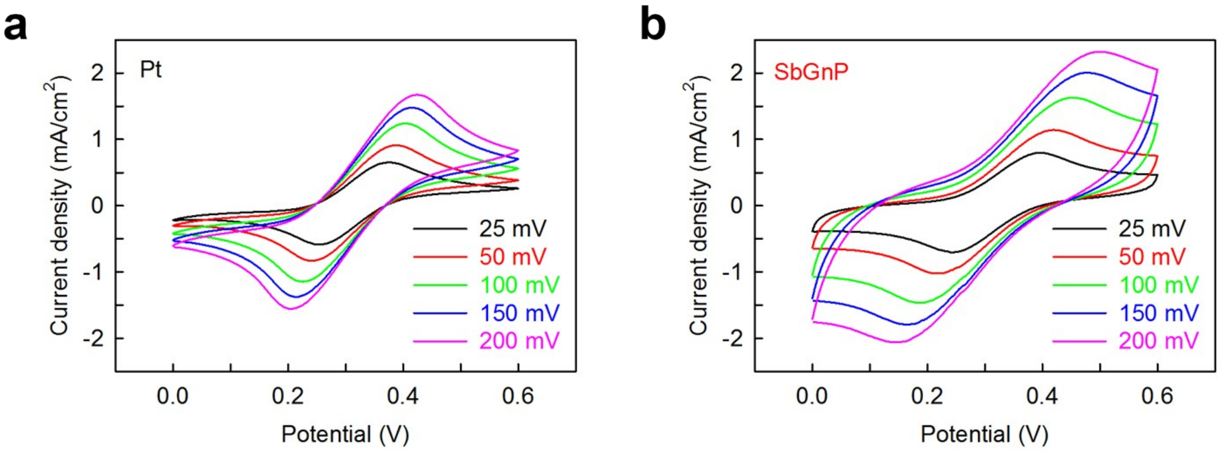

Figure S6. Cyclic voltammograms as a function of scan rate oxidation and reduction of the Co(bpy)$_3^{2+/3+}$ redox couple using Pt (a) and SbGnPs (b) electrodes as the working electrodes, a Pt wire as the CE, Ag/Ag$^+$ as the reference electrode, and 0.1 M LiClO$_4$ as the supporting electrolyte.

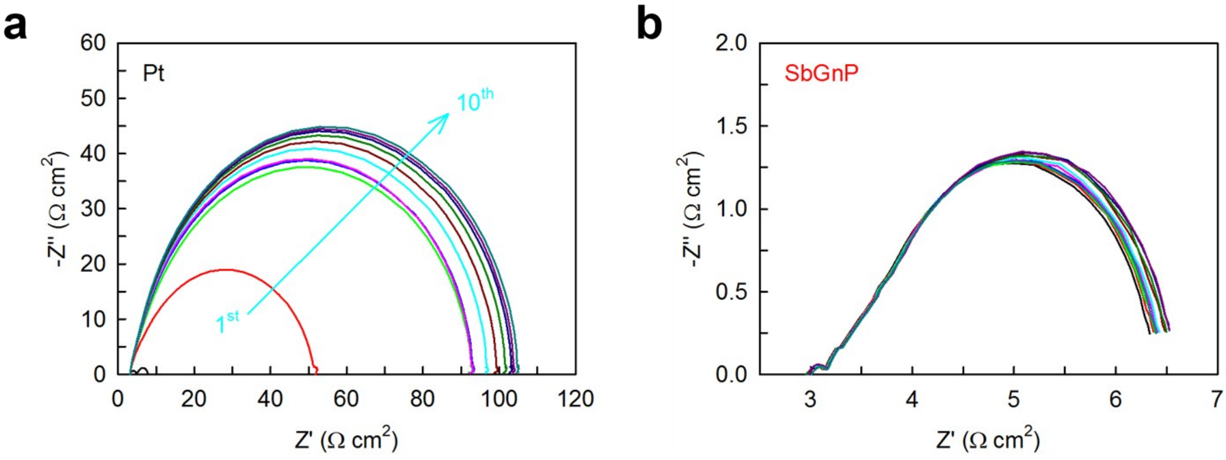

Figure S7. Nyquist plots of the symmetrical dummy cells with different CEs on the Co(bpy)$_3^{2+/3+}$ redox couple. The sequence of measurements is as follows: 100 × CV scans (from 0 V → 1 V→ −1 V→ 0 V at a scan rate of 50 mV s^{-1}) followed by 60 s of relaxation at 0 V and then EIS measurement at zero bias potential. The sequence of the electrochemical test was repeated 10 times.

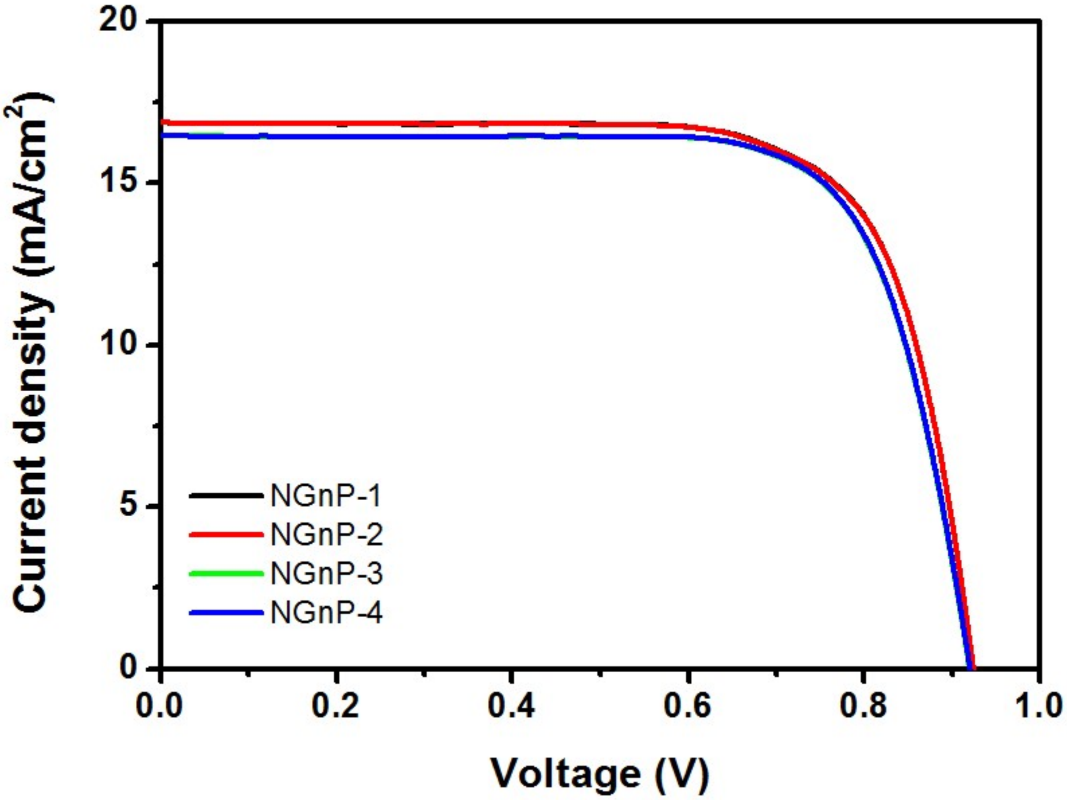

Figure S8. Current-voltage characteristics of the DSSCs with NGnP-CEs under one-sun illumination (AM 1.5G). The TiO_2 film thickness and active area are 7.0 (3.5 + 3.5) μm and 0.141 cm^2 with a black metal mask.

Table S1. EDS and XPS data of the pristine graphite and SbGnPs

Sample	Element	EDS (at.%)	XPS (at.%)
Graphite	C (%)	98.80	98.35
	O (%)	1.20	1.65
SbGnPs	C (%)	86.80	91.90
	O (%)	5.85	4.37
	Cl (%)	2.07	1.60
	Sb (%)	5.28	2.13

Table S2. BET surface area, pore volume, and pore size of the pristine graphite and SbGnPs

Sample	Surface Area (m^2/g)	Pore Volume (mL/g)	Pore Size (nm)
Graphite	2.78	0.0016	2.27
SbGnPs	302.06	0.3191	4.23

Table S3. Photovoltaic performances of the DSSCs with NGnP-CEs under one-sun illumination (AM 1.5G).

CE	J_{sc} (mA/cm^2)	V_{oc} (mV)	FF (%)	PCE (%)	R_{ch} (Ω cm^2)
NGnP-1	16.89	923.8	73.85	11.52	54.70
NGnP-2	16.88	923.7	73.85	11.51	54.72
NGnP-3	16.49	919.6	74.67	11.32	55.77
NGnP-4	16.48	920.0	74.84	11.34	55.83

Supplementary References

[S1] H. N. Tsao, C. Yi, T. Moehl, J. -H. Yum, S. M. Zakeeruddin, M. K. Nazeeruddin, M. Grätzel, *ChemSusChem* **2011**, *4*, 591.

[S2] B. J. Song , H. M. Song , I. T. Choi , S. K. Kim , K. D. Seo , M. S. Kang , M. J. Lee , D. W. Cho , M. J. Ju , H. K. Kim, *Chem. Eur. J.* **2011**, *11*, 11115.

[S3] J. D. Roy-Mayhew, D. J. Bozym, C. Punckt, I. A. Aksay, *ACS Nano* **2010**, *4*, 6203.