Supporting Information

High-surface-area nanomesh graphene with enriched edge sites as efficient metal-free cathodes for dye-sensitized solar cells

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Supporting Tables and Figrues

Table S1 Performance of pure graphene-based CEs in DSSCs reported in recent literatures (based on I^-/I_3^- redox couple)

Counter electrode	S_{BET} ($m^2\ g^{-1}$)	Electrode thickness (μm)	PCE (%)	Reference Pt (%)	PCE_{sample}/PCE_{Pt}	Reference
CVD-grown graphene	-	-	1.43	6.65	0.215	1[1]
Functionalized graphene sheets	-	-	4.99	5.48	0.911	2[2]
3D Honeycomb-Like Structured Graphene	151	-	7.80	8.00	0.975	3[3]
Graphene nanosheets	-	10	6.81	7.59	0.897	4[4]
Thermally exfoliated graphene	470	-	2.82	3.37	0.837	5[5]
3D nano-foam of few-layer graphene	-	-	5.2	5.7	0.912	6[6]
3D cauliflower-fungus-like graphene	450		8.07	7.5	1.07	7[7]
Graphene nanoplatelets	600-750	-	5.00	6.89	0.726	8[8]
Reduced graphene oxide	130	-	5.57	7.58	0.735	9[9]
Graphene nanosheets	276	-	6.93	7.23	0.958	10[10]
Graphene	-	-	5.69	-	-	11[11]
Reduced graphene oxide-900	-	-	4.0	5.1	0.784	12[12]
Vertically oriented graphene film	-	30	7.63	8.48	0.900	13[13]
Gel-coated reduce graphene oxide	-	15	7.19	7.79	0.923	14[14]
NGFs	**1920**	**7.5**	**7.32**	**7.28**	**1.005**	**This work**

Table S2 Pore structure of NGFs-10, NGFs-20 and AC

Materials	S_{BET} (m^2 g^{-1})	Pore size (nm)
NGFs-10	1397	3.79
NGFs-20	1920	4.50
AC	1906	0.89

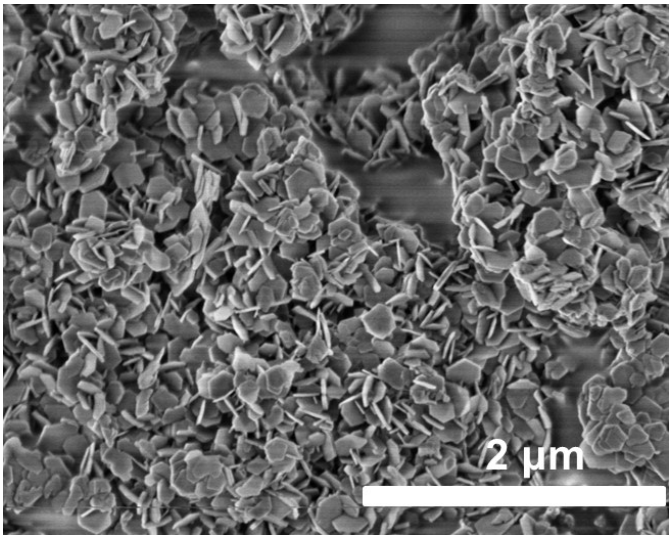

Figure S1. A SEM image of pristine $Mg(OH)_2$-derived MgO sheets.

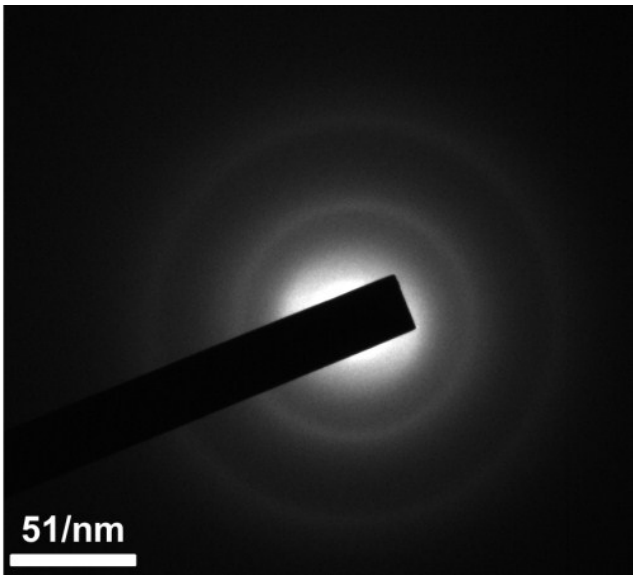

Figure S2. A electron diffraction pattern of the NGFs.

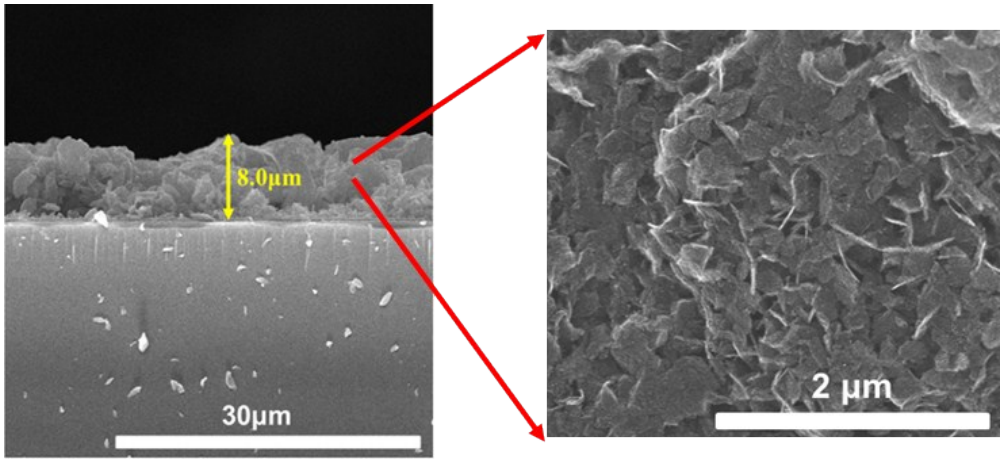

Figure S3. Cross-section SEM image of NGFs CE on FTO substrates.

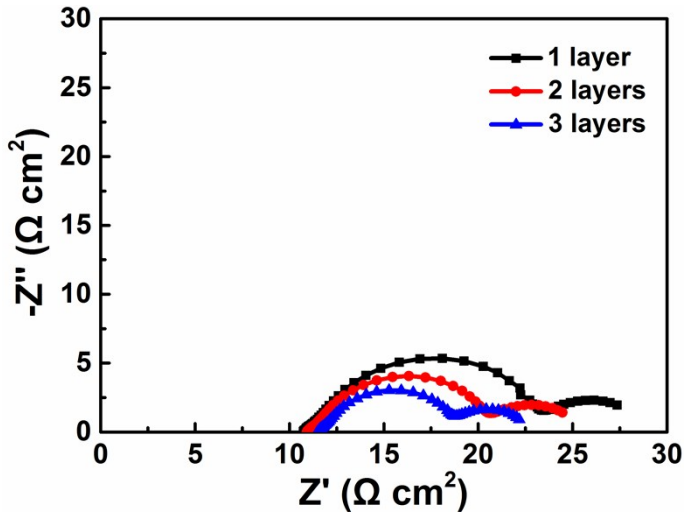

Figure S4. Nyquist plots of the symmetrical cells based on NGFs CEs with different loading.

In this work, the CE is fabricated from a slurry by screen printing. Therefore, the thicknesses of the catalysts can be easily controlled by varying the screen printing times. For all of the tested electrodes, the process of the fabrication is identical. So the thicker films correspond to higher loading of catalysts. The catalytic activity of various loading of catalyst have been measured using the impedance spectra for NGFs symmetric cells and the corresponding curves are shown in Fig. S4. It is noted that the charge transfer resistance (R_{ct}) corresponding to the catalytic activity can be obviously affect by the amount of loading nanomesh graphene.

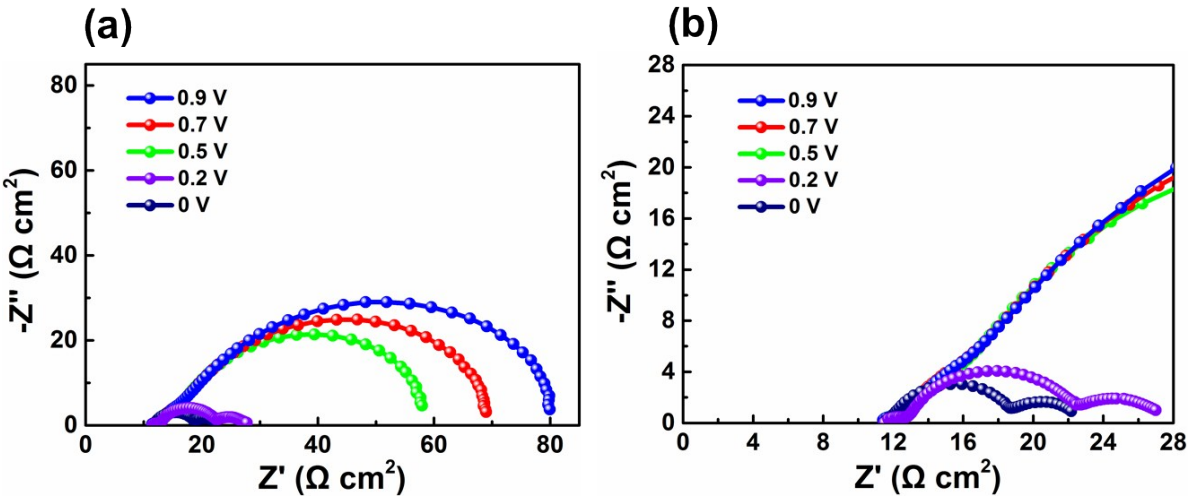

Figure S5. (a) Nyquist plot of impedance spectra on symmetrical dummy cell with NGFs electrodes at various applied biases. (b) An expansion of the high frequency region. The legend in (b) is for (a) as well.

DFT calculations

Based on our DFT calculations, the geometrical and electronic structure of graphene discussed in this work is investigated by modeling regular and defected edges. Five configurations are optimized at the theoretical level and characterized as stable points. Among them, defected edges **1-3** may have different spin states. They are singlet (**S**, without unpaired electron), triplet (**T**, with one pair of unpaired electrons in same spin state) and other high spin states. Here, we discuss only computation results of the most stable isomers of **1-3**, namely **1T**, **2T** and **3S**. The triplet **1T** has a configuration composed of 6-member-ring only, which shows the shortest C-C distance 3.121 Å. When the outside edge is formed from 5-member- and 7-member-ring only, a width of the gap ranging from 3.501 to 3.503 Å (**2T**) is calculated, very close to 3.3 Å measured in experiment (ESI, Fig. S6†). On the contrary, **3S** is the most stable configuration for the combination of 5-member- and 6-member-ring, however obviously bent as show in Fig S4. Its gap width of 4.706 Å approaches extremely the experimental measurement (5 Å) (ESI, Fig. S7†). Thus, it is far more likely that these configurations **2T** and **3S** can describe the edge structures of graphene analyzed in experiment.

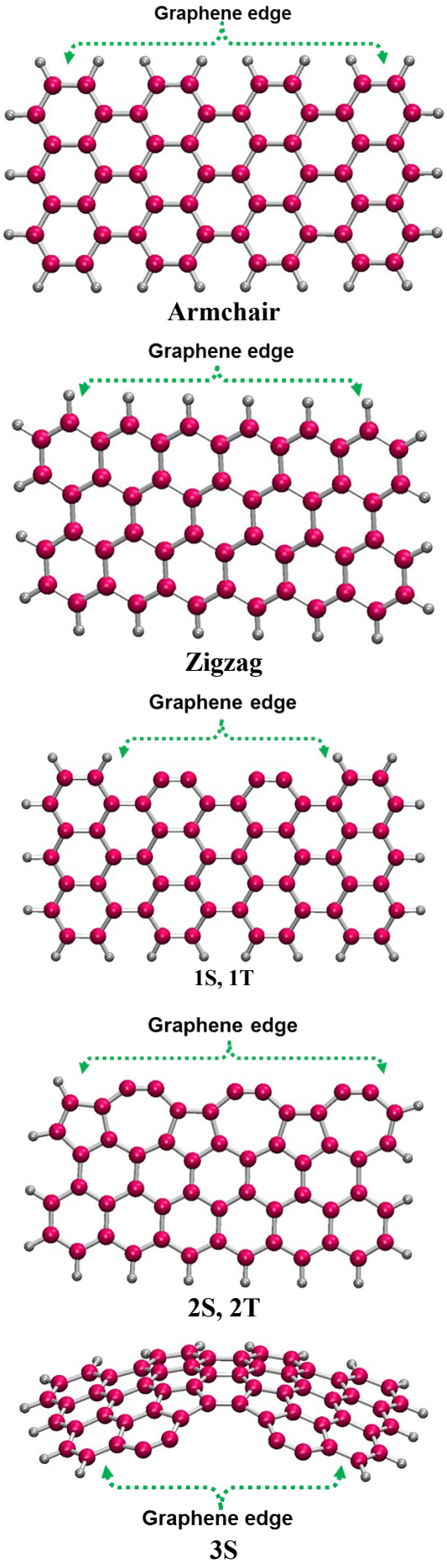

Figure S6. Five possible conformers of graphene edge.

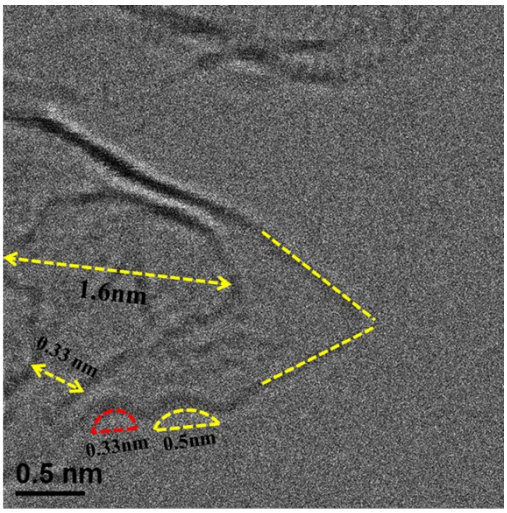

Figure S7. A high-angle annular dark field (HAADF) image of NGFs.

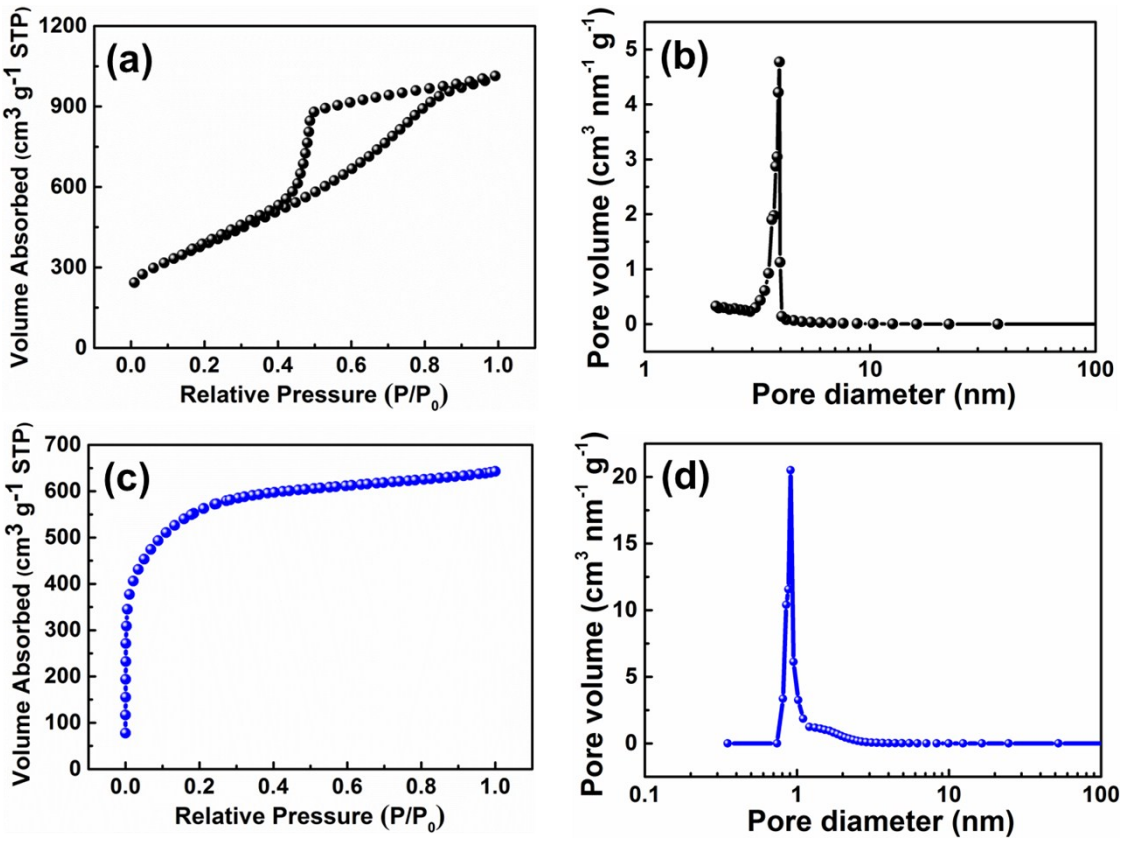

Figure S8. (a,c)Nitrogen sorption isotherms of NGFs-10 and AC, respectively. (b,d) The pore-size distribution of NGFs-10 and AC, respectively.

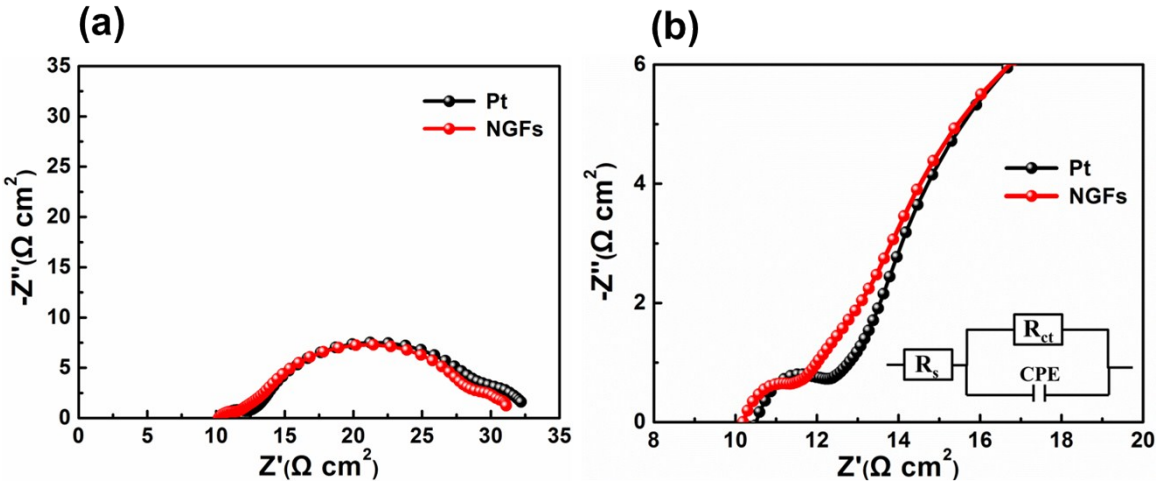

Figure S9. Nyquist plots in the full-frequency range (a) and high-frequency region (b) of complete DSSCs with Pt and NGFs CEs under one-sun illumination. Equivalent circuit for fitting at the high-frequency region is given in the inset.

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