Supporting Informantion

Graphene Coated Nanoporous Nickel towards Metal-Catalyzed Oxygen Evolution Reaction

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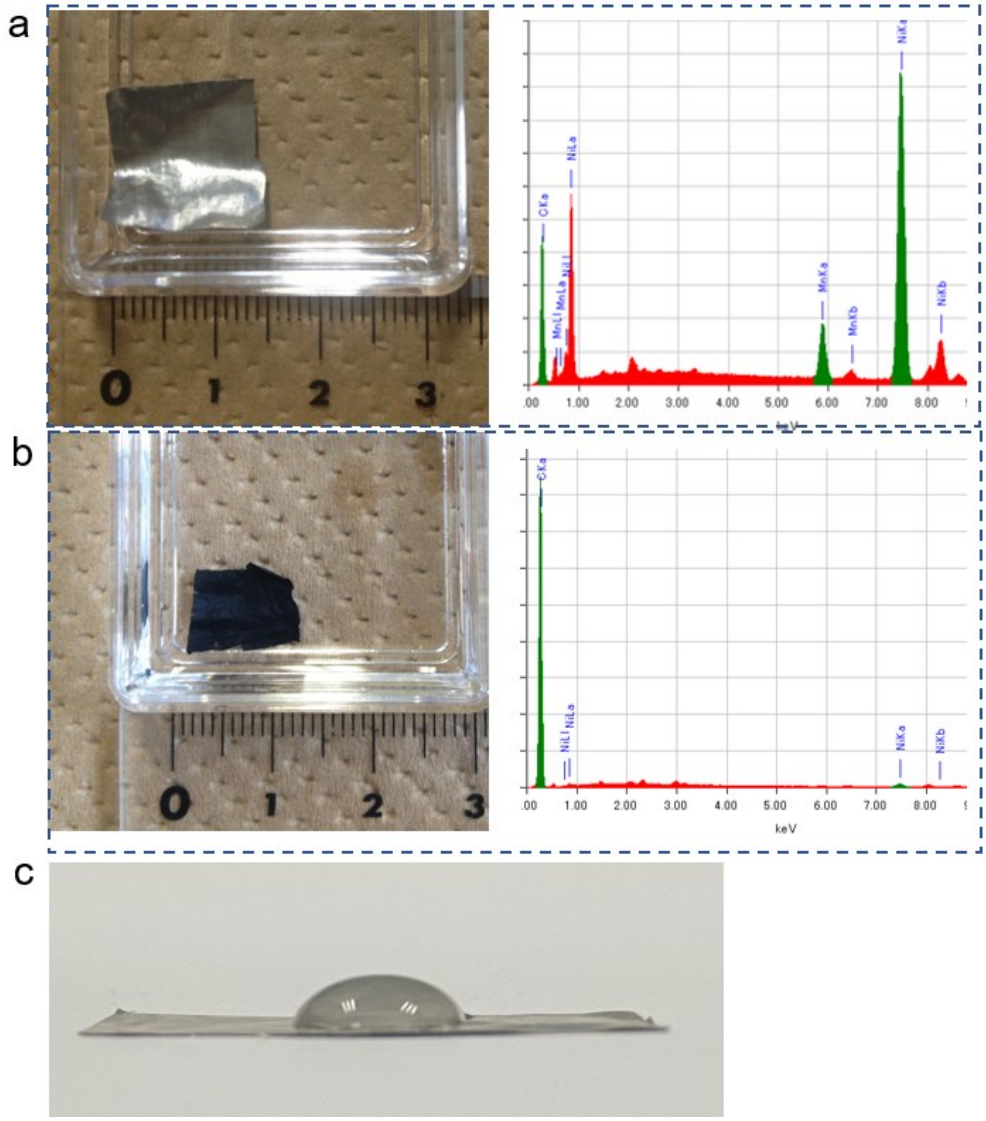

Figure S1. Photos and EDS spectra of graphene@np-Ni before (a) and after (b) dissolution of Ni. The scale in the ruler is centimeter. From the EDS spectrum of graphene@np-Ni shown in Figure S1, a small amount of Mn (~10 at.%) can also be detected. The existence of some active component (Mn) in dealloyed nanoporous metal is a normal phenomenon because the dealloying process cannot completely remove Mn from the precursor NiMn binary alloy. (c) Photograph of water droplet contact angle showing the good wettability of the graphene@np-Ni composite.

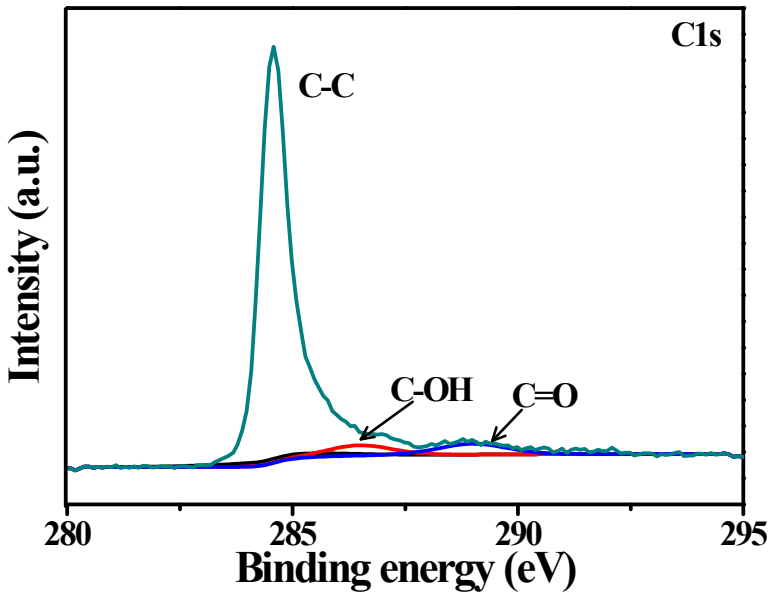

Figure S2. XPS spectrum of C1s of the graphene coatings.

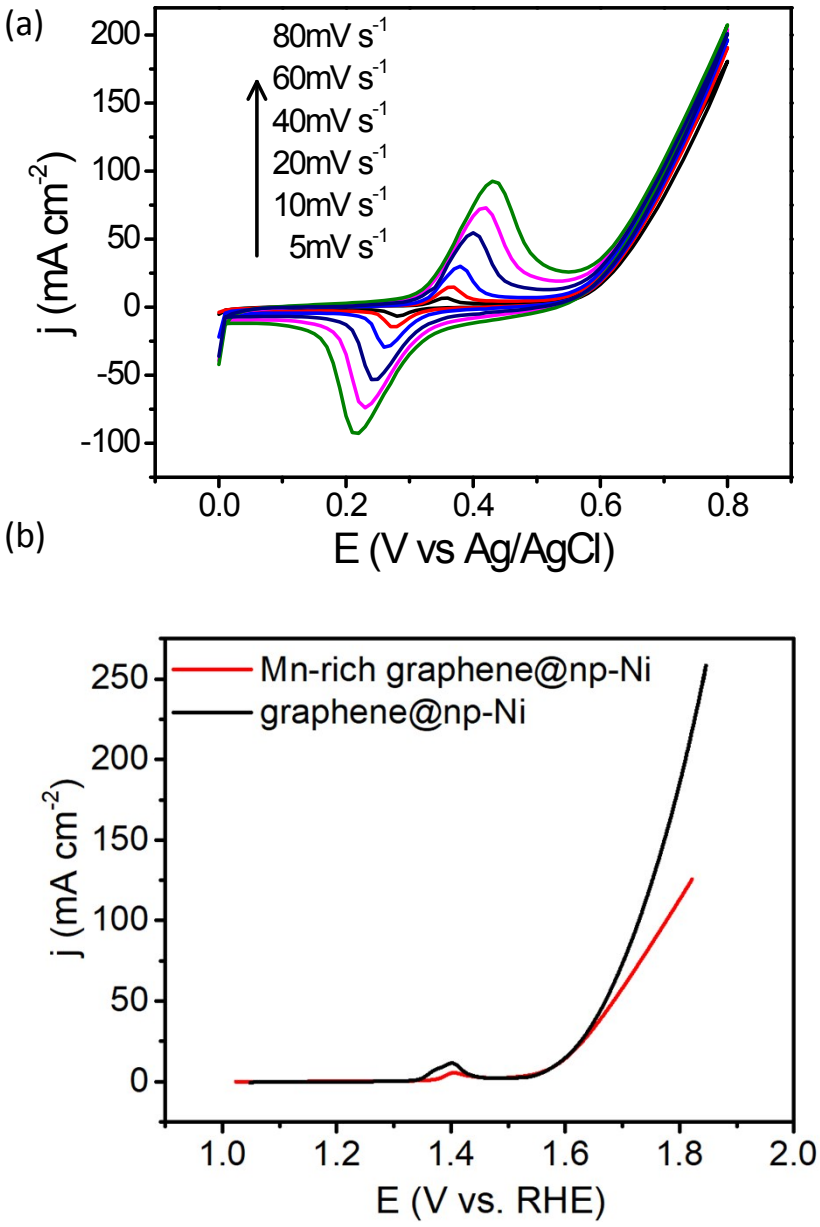

Figure S3. CV curves of the graphene@np-Ni with different scan rates (a). LSV curves of the graphene@np-Ni with residual less Mn (black) and with more Mn (red, b). The Mn-rich sample was obtained by dealloying with less time and the Mn content is around 20 at.% compared with the residual 10 at.% of our original sample. It was found that with more Mn, the graphene@np-Ni shows clearly decrease OER activity, suggesting Mn is not beneficial for the OER reaction.

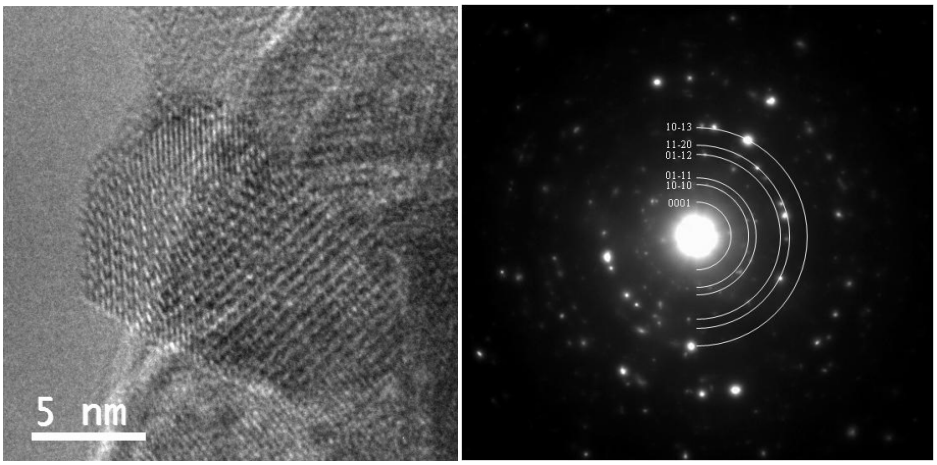

Figure S4. HRTEM image and electron diffraction pattern of the Ni hydroxide formed during OER.

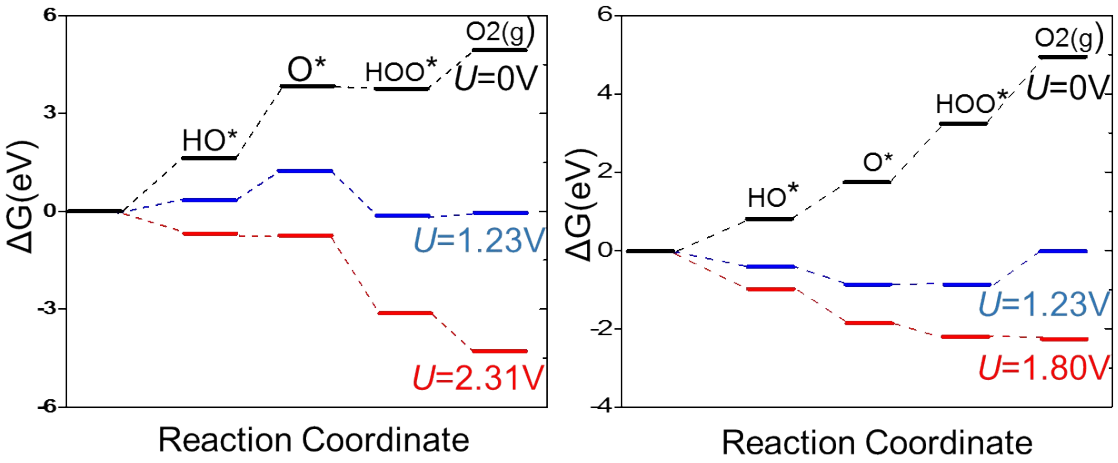

Figure S5. The OER reaction on the surfaces of (a) pristine graphene and (b) graphene@np-Ni at different potentials. All free energies are calculated at 1/8th of monolayer graphene covered with HO, O and HOO, respectively.

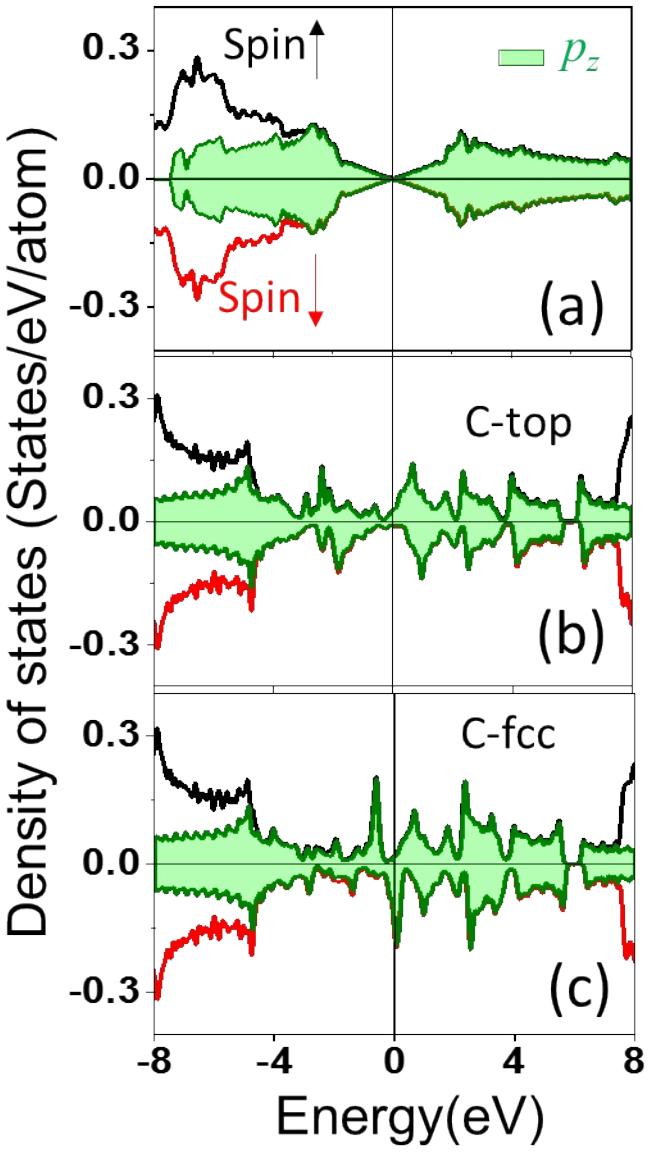

Figure S6. The projected density of states for C-atom (a) in the pristine graphene; (b) and (c) in the graphene@np-Ni systems. The inset are shown as the p_z states of C-atom.

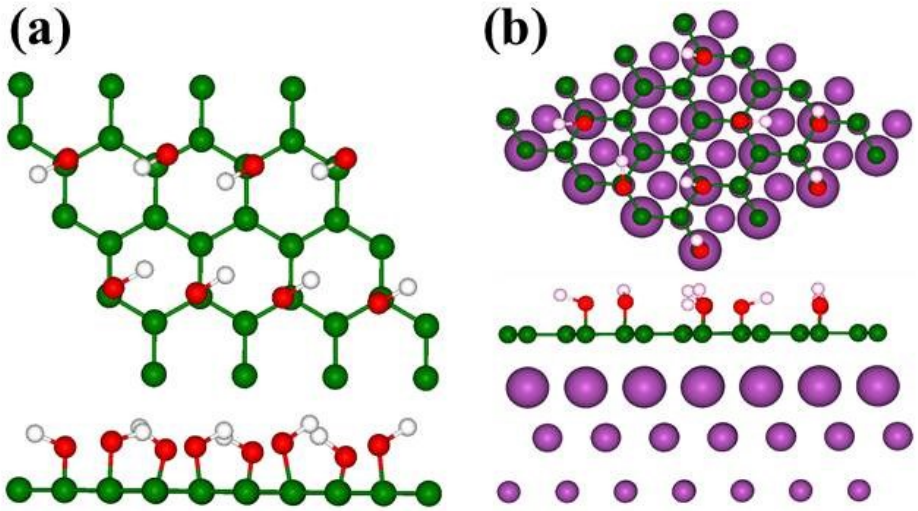

Figure S7. (a) The periodic supercell containing 8 HO molecule absorbed on the top of a 32-cites graphene slab. (b) HO absorption structure on graphene@np-Ni (111) with a computed separation of 2.17 Å. The supercell contains 48 Ni atoms for the three-layer model of the electrode (three layers of 4 × 4 Ni (111)) and a slab of 32 C atoms. Top view (top) and side view (bottom); white, red, green and purple balls denote H, O, C and Ni ions, respectively.

Table S1. Comparison of the OER performance with reported data.

Catalyst	Electrolyte	Current density （mA cm^{-2}）	Overpotential （mV）	Tafel slope (mV/decade)	Ref.
graphene@np-Ni	1.0 M KOH	50	460	45	**This work**
Fe-MOF-800	0.1 M KOH	50	~470	119	*ACS Appl. Mater. Interfaces* **2020**, 12, 40, 44710–44719
Co-N_x/C NRA	0.1 M KOH	50	~500	62.3	*Adv. Funct. Mater.* **2018**, 28, 1704638.
Co_9S_8/CNT	0.1 M KOH	50	~430	58	*J. Mater. Chem. A **2017**, 5, 21353*
Co/NGC-3	0.1 M KOH	40	~450	92	*ACS Appl. Mater. Interfaces **2020**, 12, 5, 5717–5729*
Fe_3C-Co/NC	1.0 M KOH	50	~420	not given	*Adv. Funct. Mater.* **2019**, 29, 1901949
Mn/Co-N-C	0.1 M KOH	20	~540	145	*ACS Sustainable Chem. Eng. **2019**, 7, 14180.*
$ZnCo_2O_4$-CNT	0.1 M KOH	30	~510	70.6	*Adv. Mater. **2016** 28. 3777-3784*
NCN-1000-5	0.1 M KOH	10	410	142	*Energy Environ. Sci. **2019**,12, 322-333*

Table S2. Absorption energies for reaction steps at 1/4th and 1/8th of a monolayer graphene with HO, O and HOO, respectively. A correction is added to the binding energy to obtain the free energy of 0.05, 0.35 and 0.40 eV, respectively. The free energy of the total reaction is fixed at the experimental value of 2.46 eV per water molecule, in order to avoid the calculation of the O_2 bond energy which is hard to calculate accurately with DFT.

Surfaces	Coverage	ΔE_{HO^*}	ΔE_{O^*}	ΔE_{HOO^*}
Graphene	1/8	1.26	3.85	3.39
	1/4	1.38	3.91	3.40
Graphene@np-Ni	1/8	0.49	1.73	1.83
	1/4	0.58	2.04	2.95