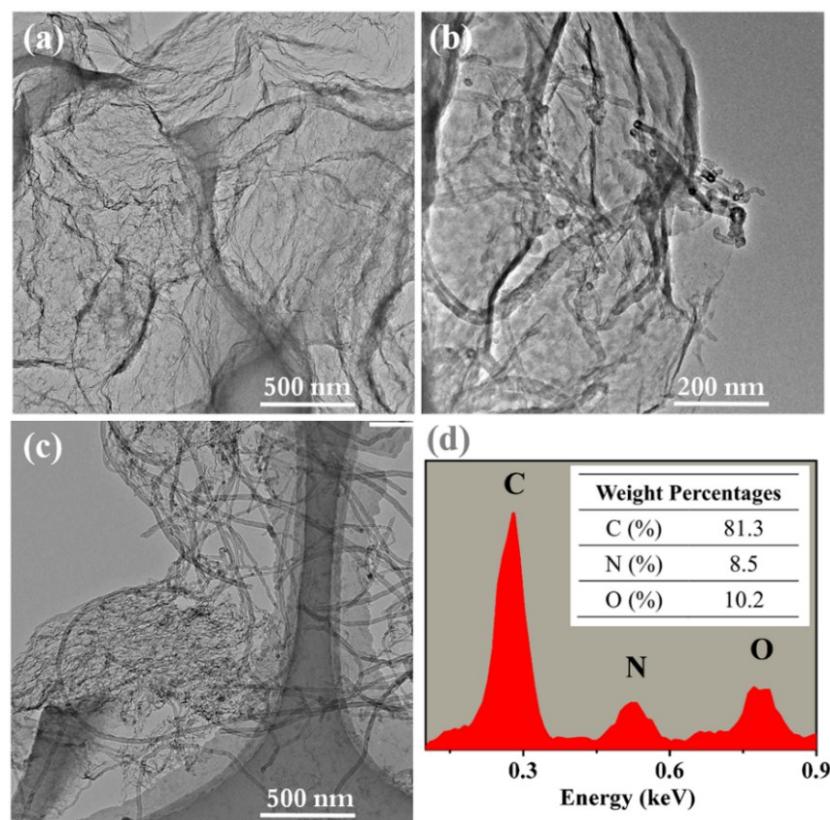


## Supporting Information for

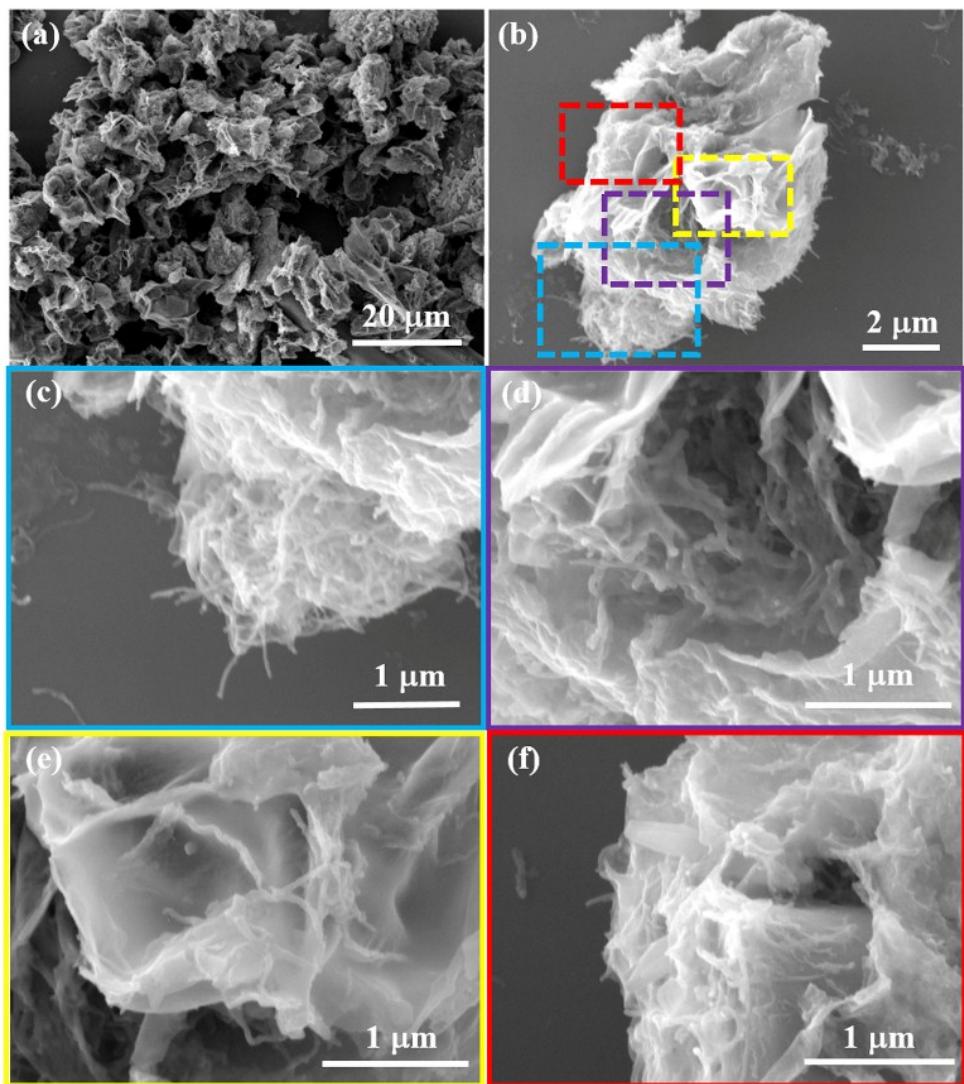
### **Nitrogen-Doped Mesoporous Carbon Nanosheet/Carbon Nanotube Hybrids as Metal-Free Bi-functional electrocatalysts for Water Oxidation and Oxygen Reduction**

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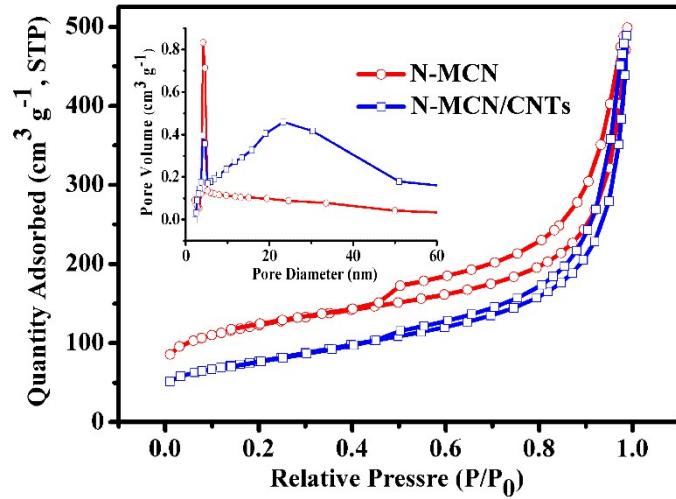
State Key Laboratory of Applied Organic Chemistry, The Key Laboratory of Catalytic Engineering of Gansu Province and Chemical Engineering, College of Chemistry and Chemical Engineering, Lanzhou University, Lanzhou, 730000, P. R. China. \*E-mail:  
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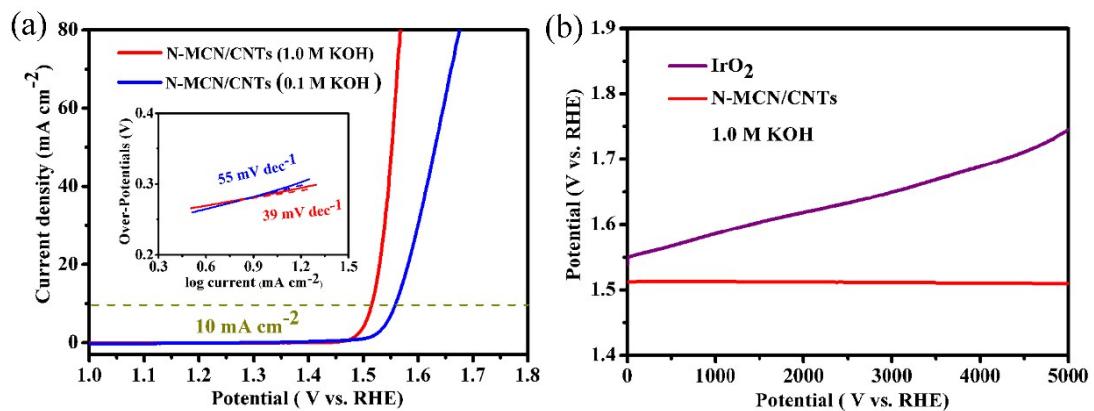
**Fig. S1** TEM images of (a) N-MCN, (b) N-MCN/CNTs, (c) mixed N-MCN+CNTs, and (d) EDX characterization.



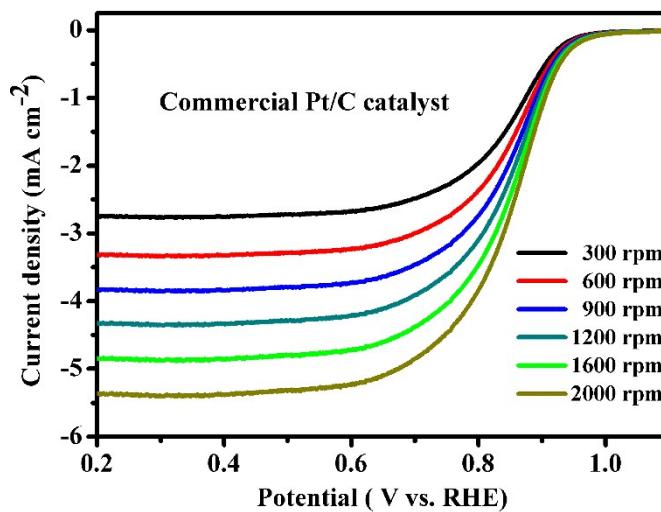
**Fig. S2** SEM images to show the homogeneity in morphology and its structure of N-MCN/CNTs (images c-f are local image of b).



**Fig. S3** Nitrogen adsorption–desorption isotherms of N-MCN and N-MCN/CNTs (inset: the corresponding pore-size distributions curves).



**Fig. S4** (a) LSVs of N-MCN/CNTs in 0.1 M and 1.0 M KOH solutions, respectively.  
 (b) Chronopotentiometric response of N-MCN/CNTs in 1.0 M KOH solution as compared to that obtained for  $\text{IrO}_2$  catalyst.



**Fig. S5** LSVs of commercial 20% Pt/C catalyst in O<sub>2</sub>-saturated 0.1M KOH with a sweep rate of 5 mV s<sup>-1</sup> at the different rotation rates.

**Table S1** XPS results of N-MCN/CNTs on the atomic percentages of C, N, O and the N distributions.

C (%)	N (%)	O (%)	N distribution (%)			
			pyridinic N	pyrrolic N	graphitic N	oxygenated N
85.1	10.7	4.2	37.7	20.5	31.6	10.2

**Table S2** Comparison of the OER activity for several recently reported high performance OER catalysts.

Catalysts	Onset potential (V)	$\eta@10.0\text{ mA cm}^{-2}$ (V)	Tafel slope (mV dec <sup>-1</sup> )	Catalyst loadings (mg cm <sup>-2</sup> )	Electrolyte	Reference
<b>N-MCN/CNTs</b>	<b>1.50</b>	<b>0.32</b>	<b>55</b>	<b>0.21</b>	<b>0.1 M KOH</b>	This work
	<b>1.46</b>	<b>0.28</b>	<b>39</b>	<b>0.21</b>	<b>1.0 M KOH</b>	This work
g-C <sub>3</sub> N <sub>4</sub> NS-CNT	1.53	0.37	83	0.20	0.1 M KOH	<sup>1</sup>
	1.47	~0.29	/	0.20	1.0 M KOH	<sup>1</sup>
N/C	~1.50	0.38	/	0.20	0.1 M KOH	<sup>2</sup>
N <sub>x</sub> P-GCNS	1.32	0.34	70	0.14	0.1 M KOH	<sup>3</sup>
NGSH	~1.44	0.40	83	0.25	0.1 M KOH	<sup>4</sup>
NPMC-1000	~1.30	~0.60	/	0.15	0.1 M KOH	<sup>5</sup>
OCC-8	~1.57	0.48	82	/	0.1 M KOH	<sup>6</sup>
g-C <sub>3</sub> N <sub>4</sub> /graphene	1.5	0.54	68.5	0.48	0.1 M KOH	<sup>7</sup>
Fe <sub>3</sub> C@NG800-0.2	~1.50	0.36	62	0.20	0.1 M KOH	<sup>8</sup>
Au@Co <sub>3</sub> O <sub>4</sub> /C	~1.51	0.35	60	0.20	0.1 M KOH	<sup>9</sup>
Ni-P	1.48	0.30	64	0.20	1.0 M KOH	<sup>10</sup>
Ni–Co ADHs nanocages	1.50	0.35	65	0.2	1.0 M KOH	<sup>11</sup>
Co-P/NC	~1.50	0.32	52	0.28	1.0 M KOH	<sup>12</sup>
N-CG–CoO	~1.52	0.34	71	0.71	1.0 M KOH	<sup>13</sup>
Co <sub>3</sub> O <sub>4</sub> @C-MWCNTs	1.50	320	62	0.33	1.0 M KOH	<sup>14</sup>
FeNi@NC	1.44	0.28	70	0.32	1.0 M NaOH	<sup>15</sup>

NiCo-LDH	~ 1.52	0.37	40	0.17	1.0 M KOH	<sup>16</sup>
Ni <sub>2</sub> P	~ 1.47	0.29	/	0.14	1.0 M KOH	<sup>17</sup>
CoOx@CN	1.51	~0.33	60	0.14	1.0 M KOH	<sup>18</sup>
CNTs-Au@Co <sub>3</sub> O <sub>4</sub>	1.52	0.35	68	0.36	1.0 M KOH	<sup>19</sup>
Zn <sub>x</sub> Co <sub>3-x</sub> O <sub>4</sub> nanowire	~ 1.50	0.32	51	1.0	1.0 M KOH	<sup>20</sup>

**Notes:** For the convenience of comparison, the measure potentials vs. Ag/AgCl were converted to a reversible hydrogen electrode (RHE) scale accorting to the Nerst equation ( $E_{\text{RHE}} = E_{\text{Ag/AgCl}} + 0.059 \times \text{pH} + 0.198$ ).

**Table S3** Comparison of the ORR activity for several recently reported high performance ORR catalysts (electro rotating speed is 1600 rpm).

Catalysts	Onset	Half-wave	Electron transfer	Catalyst loadings	Electrolyte	Reference
	potential (V)	potential (V)	numbers	(mg cm <sup>-2</sup> )		
<b>N-MCN/CNTs</b>	<b>0.95</b>	<b>0.82</b>	<b>3.8-3.9</b>	<b>0.21</b>	<b>0.1 M KOH</b>	<b>This work</b>
N-doped porous C	0.86	0.7	3.3-3.9	0.20	0.1 M KOH	<sup>21</sup>
N,P-doped C fiber	0.88	0.79	~ 4	0.10	0.1 M KOH	<sup>22</sup>
NG-NCNT	0.87	/	3.7	0.05	0.1 M KOH	<sup>23</sup>
NC900	0.83	~0.68	3.3	0.034	0.1 M KOH	<sup>24</sup>
N,S,O-OMC	0.85	~0.79	3.5	0.16	0.1 M KOH	<sup>25</sup>
CoP NCs	0.80	0.70	3.5	0.28	0.1 M KOH	<sup>26</sup>
Pt/CaMnO <sub>3</sub>	0.90	~0.79	3.9	0.09	0.1 M KOH	<sup>27</sup>

NC-NZ-13	~ 0.98	~ 0.84	~ 4	0.21	0.1 M KOH	<sup>28</sup>
meso/micro-PoPD	~ 1	0.87	3.97	0.5	0.1 M KOH	<sup>29</sup>
NC-A	~ 0.975	0.832	0.372	0.128	0.1 M KOH	<sup>30</sup>
NDCN-22	0.955	0.855	3.67–3.94	0.6	0.1 M KOH	<sup>31</sup>
NG-900	0.935	~ 0.785	3.70	0.051	0.1 M KOH	<sup>32</sup>
NG/Fe <sub>10.0</sub>	0.961	~ 0.74	3.91	0.051	0.1 M KOH	<sup>32</sup>

**Notes:** For the convenience of comparison, the measure potentials vs. Ag/AgCl were converted to a reversible hydrogen electrode (RHE) scale accorting to the Nerst equation ( $E_{\text{RHE}} = E_{\text{Ag/AgCl}} + 0.059 \times \text{pH} + 0.198$ ).

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