Supporting information for

In Situ Synthesis of Ultrathin Metal-Organic Framework Nanosheets: A New Method for 2D Metal-Based Nanoporous Carbon Electrocatalysts

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Fig. S1 Additional (a) large-area and (b) high-magnification TEM images of ZIF-67 nanosheets.



Fig. S2 (a) XRD patterns of simulated ZIF-67 and as-synthesized ZIF-67 nanosheets.(b) The crystal structure of ZIF-67, yellow represents void space and blue represent CoN₄ tetrahedra, respectively.



Fig. S3 SEM images (a-b), AFM image (c) and corresponding height profiles (d) of Co,N-C NS-800°C.



Fig. S4 SEM images of (a) Co,N-C NS encapsulated NaCl template and (b) Co,N-C NS-800°C with hollow box after NaCl template removal.



Fig. S5 TEM images of Co,N-C NS-800°C with (a) graphitic carbon nanopores and (b) ultrathin edge thickness. The interplanar crystalline space is 0.34 nm.



Fig. S6 (a) EDS spectrum and (b) TGA curve of Co,N-C NS-800°C.



Fig. S7 (a-b) TEM images of rhombic dodecahedral ZIF-67 nanocrystals. (c-e) TEM images and (f) HRTEM image of Co,N-C NP-800°C.



Fig. S8 SEM images (a), (d) and (g), and TEM images (b-c), (e-f) and (h-i) of Co,N-C NP-600°C, Co,N-C NP-700°C and Co,N-C NP-850°C, respectively.



Fig. S9 (a) Raman spectra of Co,N-C NS-600°C, Co,N-C NS-700°C, Co,N-C NS-850°C and Co,N-C NP-800°C. There are two dominant peaks at 1355 and 1580 cm⁻¹, corresponding to D and G bands, Respectively. (b) TGA curve of Co,N-C NP-800°C.



Fig. S10 XPS spectrum of Co,N-C NS-800°C



Fig. S11 High resolution Co 2p XPS spectra of (a) Co,N-C NS-600 $^{\circ}$ C, (b) Co,N-C NS-700 $^{\circ}$ C, (c) Co,N-C NS-850 $^{\circ}$ C and (d) Co,N-C NP-800 $^{\circ}$ C.



Fig. S12 (a) SEM image and (b) TEM image of Co,N-C materials with the same reaction conditions as the Co,N-C NS-800 $^{\circ}$ C except the twofold of Co²⁺ and MeIm precursors.



Fig. S13 (a) SEM image and (b) TEM image of Co,N-C materials with the same reaction conditions as the Co,N-C NS-800 $^{\circ}$ C except the 8 mL of solvent in synthesis of ZIF-67 nanosheets .



Fig. S14 Optical images of intermediate products from ZIF-67 nanosheets to Co,N-C NS-800°C. The high-yield synthetic strategy only limited by the volume of reactor and tube furnace.

Table S1. Comparisons of ORR performances for Co,N-C NS-800°C with other cobalt-
based carbon electrocatalysts in alkaline solution.

Electrocatalysts	E _{onset} (vs. RHE)	E _{1/2} (vs. RHE)	References
Co,N-C NS-800°C	0.938	0.869	This work
Co,N-CNF	0.883	0.81	[1]
Co-C@NWCs	0.939	0.83	[2]
Co3S4–S/G	0.920	0.805	[3]
cal-CoZIF-VXC72	-	0.84	[4]
Z8-Te-1000	0.825	0.74	[5]
Co/N-CNT	0.940	0.84	[6]
Co ₃ O ₄ /N-rmGO	0.880	0.83	[7]
Co@N-CNTs	0.929	0.849	[8]
Co ₄ N/CNW/CC	-	0.80	[9]
NCNTFs	-	0.87	[10]
Co/NC	-	0.83	[11]
CS-FePC_450	0.970	0.87	[12]
Fe-N-SCCFs	-	0.88	[13]
N-MC/rGO-800	0.960	0.82	[14]
Co-Mo-C/NRGO	0.869	0.786	[15]

*NS: nanosheets; NF: nanoframework; WCs: wrinkles carbon nanosheets; CNT: carbon nanotubes; CC: carbon cloth; CNTFs: carbon nanotube frameworks.

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