

Figure S1. XPS of C 1s

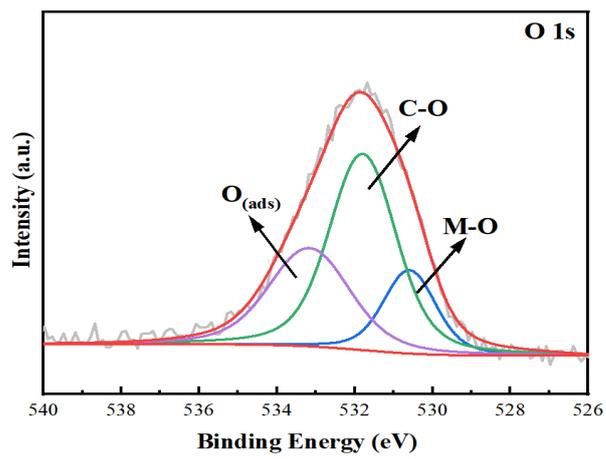
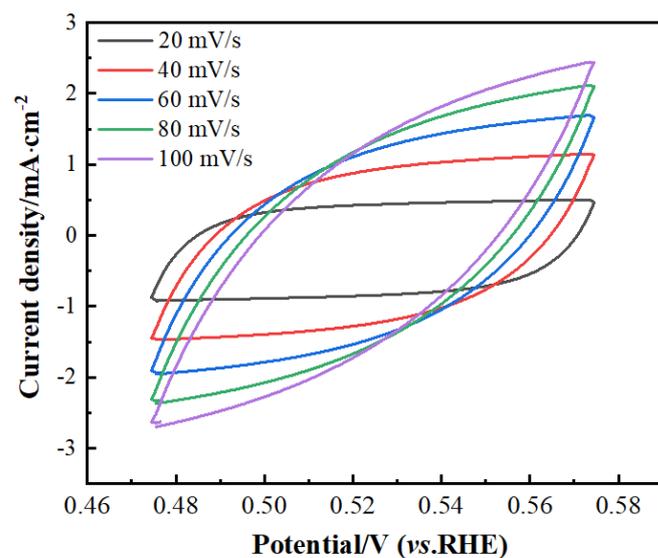
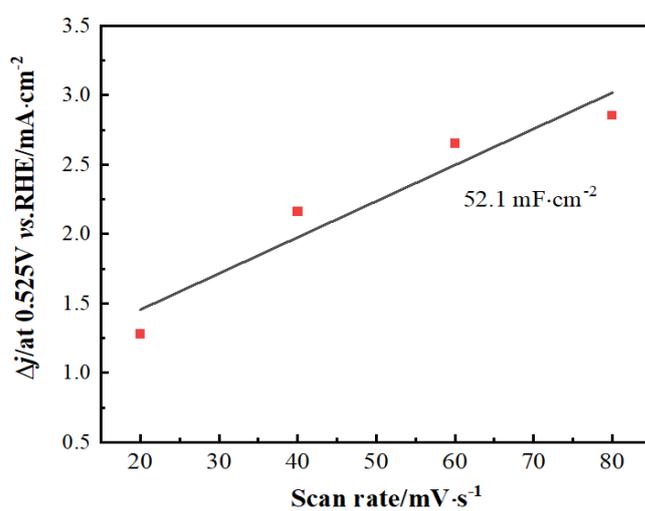


Figure S2. XPS of O 1s

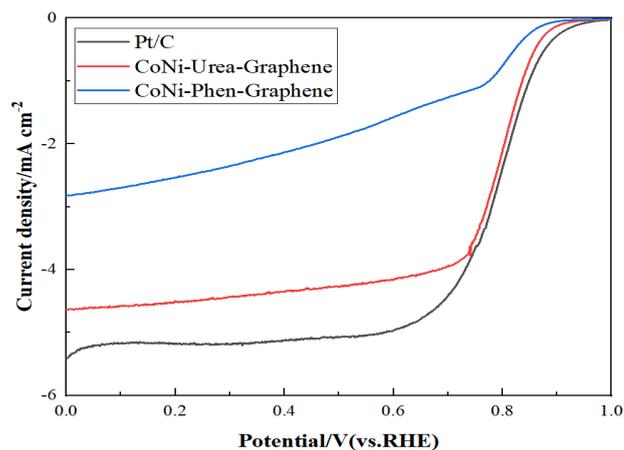


**Figure S3.** CV curves of CoNi/graphene at different sweep speeds in the range of 0.46V-0.58V

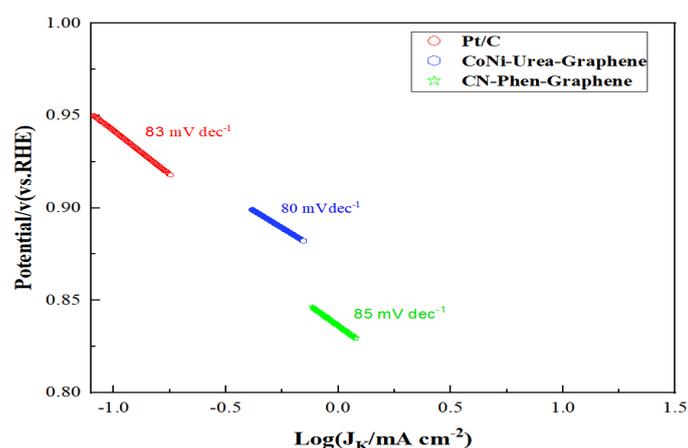


**Figure S4.** Based on the CV curve, the relationship between current density and sweep speed at 0.525V

Only Phen and only urea are added to the catalyst as reference catalysts, marked as CoNi-Phen-Graphene, CoNi-Urea-Graphene.



**Figure S5.** LSV curves of CoNi-Urea-Graphene, CoNi-Phen-Graphene and Pt/C for ORR recorded at  $5.0 \text{ mV s}^{-1}$  and 1600 rpm on RDE in  $\text{O}_2$ -saturated  $0.1 \text{ mol L}^{-1}$  KOH,



**Figure S6.** Corresponding Tafel plots

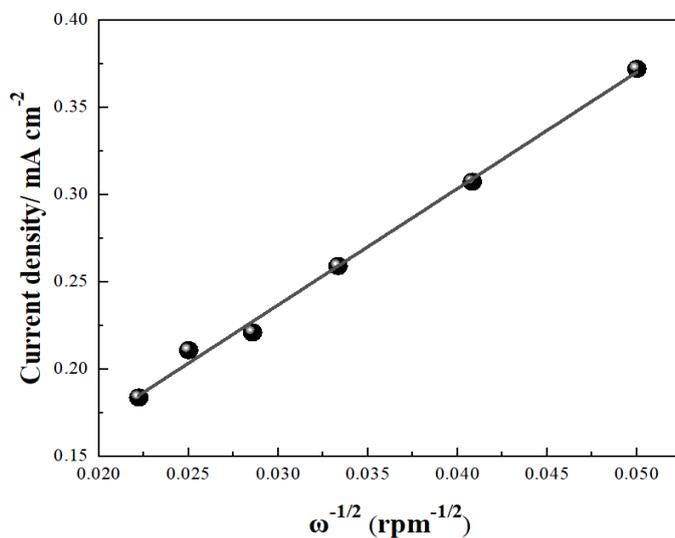
The number of electron transfer ( $n$ ) of each  $\text{O}_2$  molecule during ORR can be calculated by the slope of Koutecky–Levitch (K–L) as the following equation:

$$\frac{1}{j} = \frac{1}{j_L} + \frac{1}{j_k} = \frac{1}{B\omega^{1/2}} + \frac{1}{j_k}$$

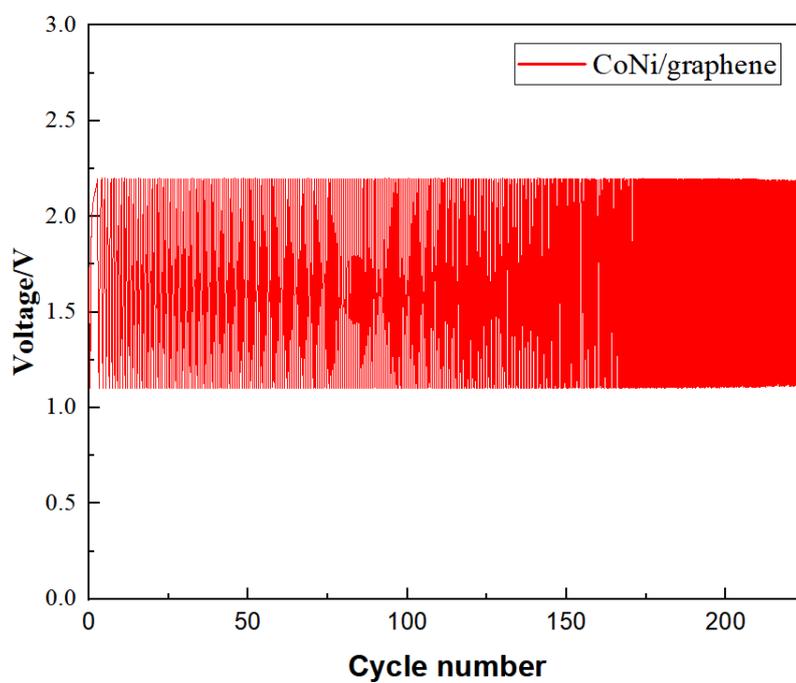
$$B = 0.2nFC_0D_0^{2/3}\nu^{-1/6}$$

where  $j$  is the measured current density,  $j_k$  is the kinetic current density, and  $j_L$  is the limiting diffusion current density.  $B$  is the Levitch constant,  $F$  is the Faraday constant ( $96485 \text{ C mol}^{-1}$ ),  $\omega$  is the rotational speed (rpm),  $C_0$  is the volume concentration of saturated oxygen in  $0.1 \text{ M KOH}$  ( $1.2 \times 10^{-6} \text{ mol cm}^{-3}$ ),  $D_0$  is the diffusion coefficient of

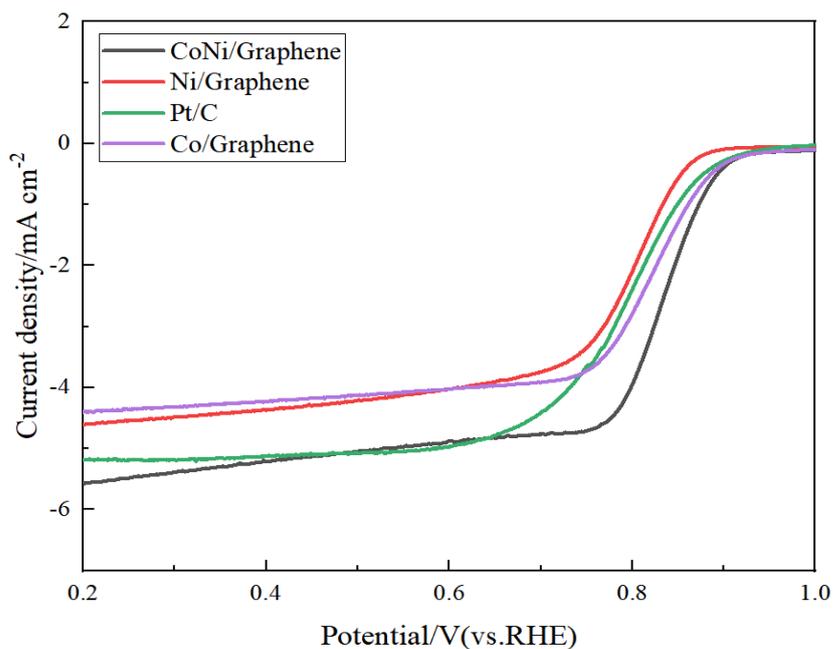
oxygen in the electrolyte solution ( $1.73 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$ ),  $\nu$  is the vis-cosity coefficient ( $0.01 \text{ cm}^2 \text{ s}^{-1}$ ) in the 0.1 M KOH electro-lyte solution, and  $n$  is the electron transfer number.



**Figure S7.** Electron transfer numbers ( $n$ ) of ORR on the CoNi/graphene

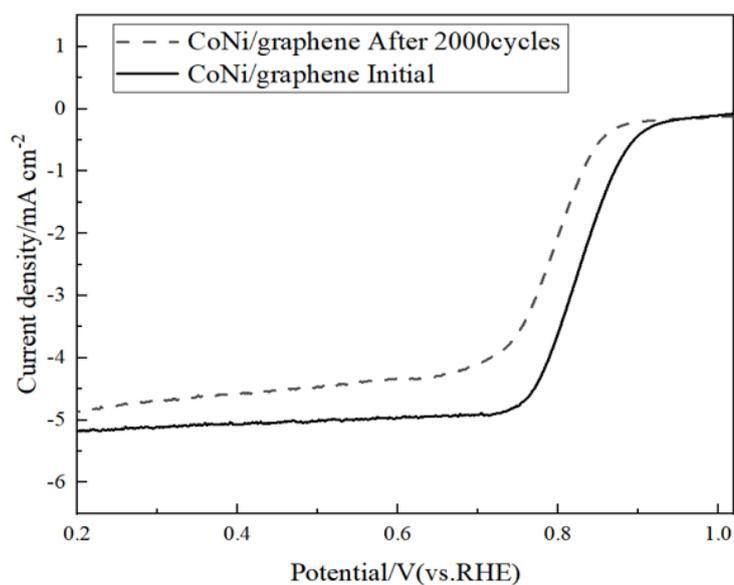


**Figure S8.** Galvanostatic discharge and charge cycling curves of SZABs with 15 min discharge and 15 min charge at  $8 \text{ mA cm}^{-2}$



**Figure S9.** LSV curves of CoNi/Graphene, Co/Graphene, Ni/Graphene and Pt/C for ORR recorded at  $5.0 \text{ mV s}^{-1}$  and 1600 rpm on RDE in  $\text{O}_2$ -saturated  $0.1 \text{ mol L}^{-1} \text{ KOH}$

*(In the contrast catalyst, only Co, Ni and CoNi were added, and the other nitrogen and carbon sources did not change.)*



**Figure S10.** ORR polarization curves at 1600 rpm with a scan rate of  $10 \text{ mV s}^{-1}$  before and after 2000 cycles accelerated degradation test

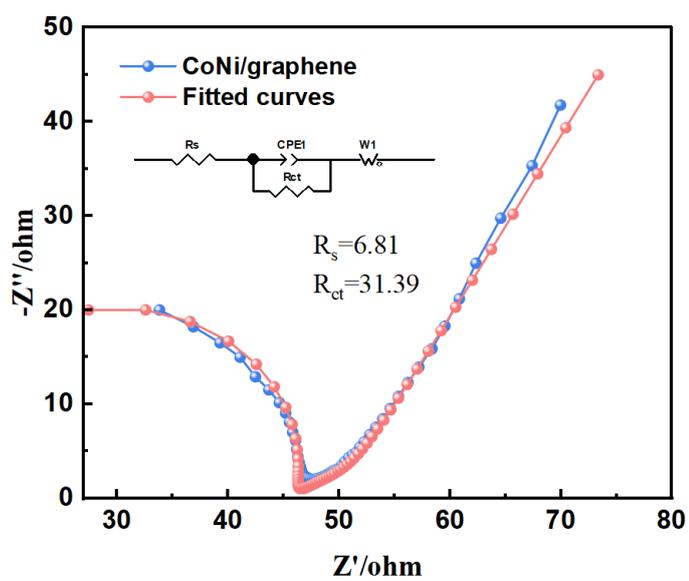


Figure S11. electrochemical impedance spectroscopy of CoNi/graphene

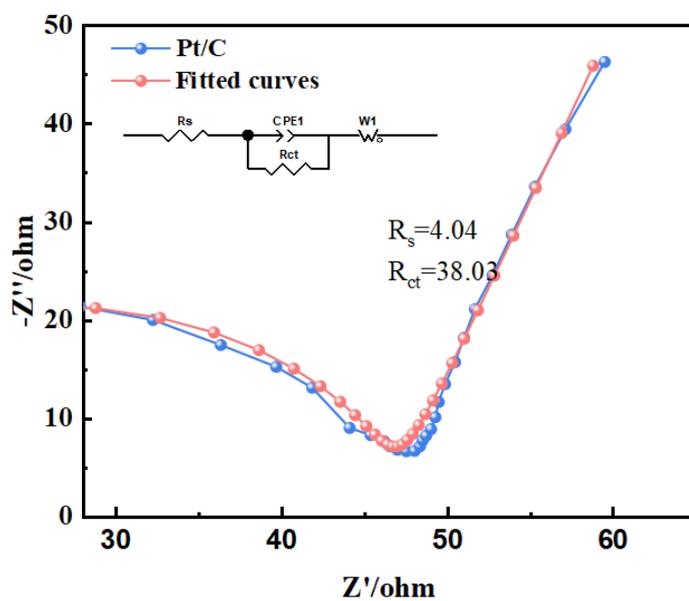
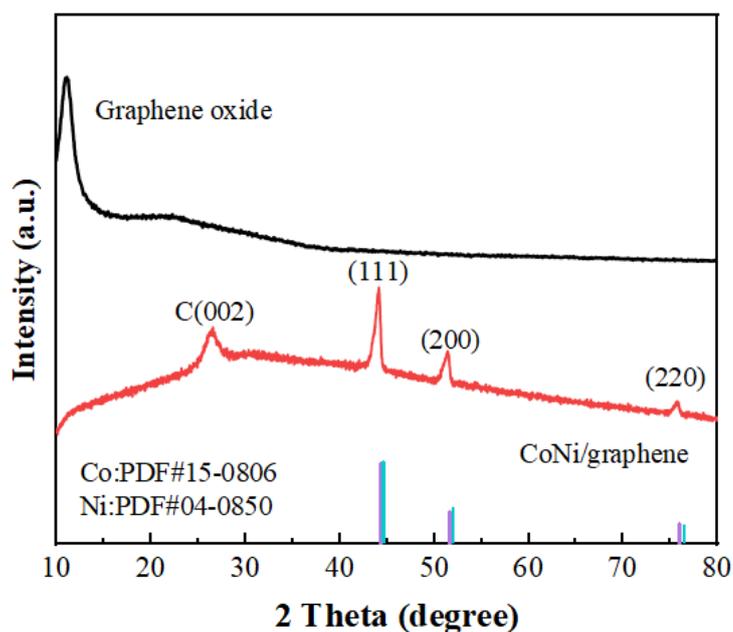


Figure S12. electrochemical impedance spectroscopy of Pt/C



**Figure S13.** XRD patterns of Graphene oxide and CoNi/graphene

**Table S1.** Comparison of ORR performance for CoNi/graphene with other non-precious metal electrocatalysts in alkaline media

Catalyst	Electrolyte	Half-wave Potential (V vs. RHE)	Limiting Current Density @1600 rpm ( $\text{mA cm}^{-2}$ )	Reference
N-doped carbon nanotube arrays	0.1 M KOH	0.84	5.6	Science 2009, 323, 760
Fe-N-C capsules	0.1 M KOH	0.83	4.9	ACS Nano 2016, 10, 5922
N-doped porous carbon fiber	0.1 M KOH	0.82	4.7	Adv. Mater. 2016, 28, 3000
N, S-doped Graphene sheets	0.1 M KOH	0.77	4.4	Nano Energy 2016, 19, 373

FeNi/N-LCN	0.1 M KOH	0.85	4.8	Nano Letter 2021, 21, 3105
N, S-doped porous carbo	0.1 M KOH	0.85	5.8	Energy Environ. Sci. 2017, 10, 742
CoNi-CoN <sub>4</sub> -HPC-900	0.1 M KOH	0.78	7.1	Nano Energy 2022, 99, 107325
CoNi/graphene	0.1 M KOH	0.83	5.7	This Work

**Table S2.** Comparison of ORR performance for CoNi/graphene with other non-precious metal electrocatalysts in alkaline media

Catalyst	Electrolyte	Half-wave Potential (V vs. RHE)	Limiting current density @1600 rpm (mA cm <sup>-2</sup> )	Power density (mW cm <sup>-2</sup> )	Reference
Fe-N-C capsules	0.1 M KOH	0.83	4.9	125 (liquid state)	ACS Nano 2016, 10, 5922
N-doped porous carbon fiber	0.1 M KOH	0.82	4.7	185 (liquid state)	Adv. Mater. 2016, 28, 3000
N, S-doped Graphene sheets	0.1 M KOH	0.77	4.4	--	Nano Energy 2016, 19, 373
CoDNi-N/C	0.1 M KOH	0.84	5.5	--	Appl. Catal. B Environ. 2019, 112 - 121
FeNi/N-LCN	0.1 M KOH	0.85	4.8	162 (liquid state)	Nano Letter 2021, 21, 3105
Co/N@CNTs@CN	0.1 M	0.86	5.5	133	Adv. Funct.

MF	KOH			(liquid state )	Mater. 2020, 2003407
CoNi-CoN <sub>4</sub> -HPC-900	0.1 M KOH	0.78	7.1	--	Nano Energy 2022, 99, 107325
N-GQDs/NiCo <sub>2</sub> S <sub>4</sub> /C	0.1 M KOH	--	--	26.2 (solid state)	Small 2019, 15, 1903610
Co <sub>3</sub> O <sub>4</sub> @LaNiO <sub>3</sub> /NCNT	0.1 M KOH	--	--	28.2 (solid state)	Adv. Mater. 2015, 27, 5617
N@CNTs@CNMF	0.1 M KOH	--	--	26.5 (solid state)	Adv. Funct. Mater. 2020, 30, 2003407
CoNi/graphene	0.1 M KOH	0.83	5.7	210 (liquid state ) 40 (solid state)	This Work