Tannic Acid-Derived Metal-Phenolic Networks with Dual-Atom ORR and Single-Atom OER Sites for Bifunctional Oxygen Electrocatalysts

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Electrochemical measurements

The ORR electrochemical tests were conducted in a three-electrode system using the Gamry reference 600 workstations (Gamry, USA) in 0.1 M KOH. The Hg/HgO electrode, carbon rod, and glassy carbon electrode (5 mm diameter) were used as the reference electrode, counter electrode, and working electrode, respectively. 5 mg of catalyst and 5 mg of KB were dispersed in 100 μL Nafion solution and 900 μL ethanol to form a homogenous ink assisted by stirring. After that, 5 μL of the prepared ink was dropped on the surface of the RDE electrode with a loading of 0.255 mg cm^{-2}. Pt/C was maintained with the same loading of 0.255 mg cm^{-2}.

The OER electrochemical tests were conducted in a three-electrode system using the Gamry reference 600 workstations (Gamry, USA) in 1 M KOH. The Hg/HgO electrode, carbon rod, and the carbon cloth (1×1 cm^2) were used as the reference electrode, counter electrode, and working electrode, respectively. 5 mg of catalyst and 5 mg of KB was dispersed in 100 μL Nafion solution and 900 μL ethanol to form a homogenous ink assisted by stirring. After that, 100 μL of the prepared ink was dropped on the carbon cloth with a loading of 0.5 mg cm^{-2}. Pt/C+RuO_2 was maintained with the same loading of 1.0 mg cm^{-2}.

Zn-air battery measurements

The performances of the Zn-air battery were evaluated by a battery test system (LANHE CT2001A) at room temperature (25 ± 2 °C). The Zn-air battery was assembled using a polished Zn plate (thickness 0.5 mm) as the anode and the hydrophobic carbon cloth coated with $Co_{0.85}Fe_{0.15}$@TA-Salphen was used as the air cathode with a loading of 1 mg cm^{-2}.

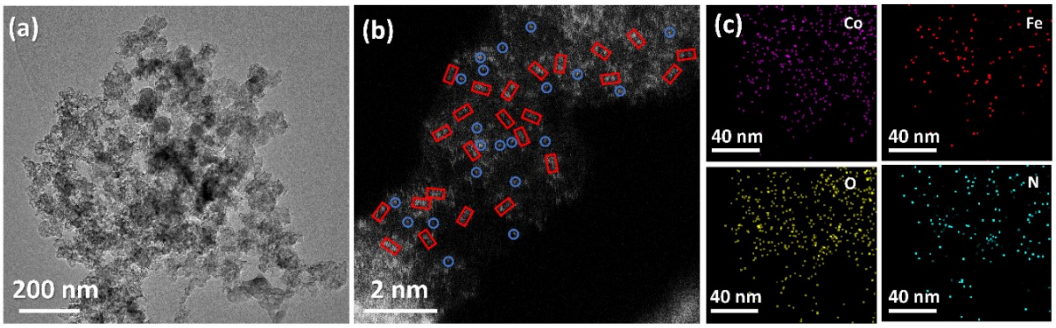

Figure S1. a, b) TEM and AC-HAADF-STEM images of $Co_{0.85}Fe_{0.15}$@TA-Salphen after stability test. c) EDS mapping of $Co_{0.85}Fe_{0.15}$@TA-Salphen after stability test.

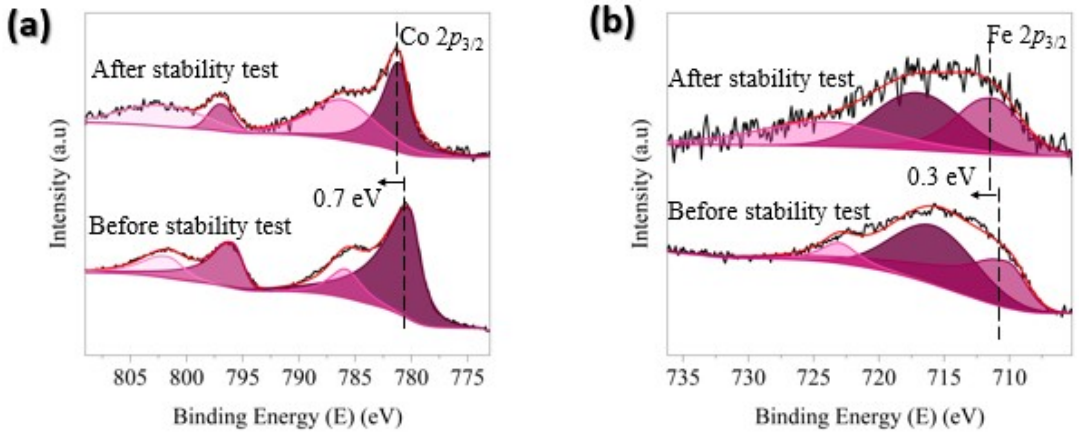

Figure S2. a) High resolution Co 2*p* and b) Fe 2*p* spectra for $Co_{0.85}Fe_{0.15}$@TA-Salphen after stability test.

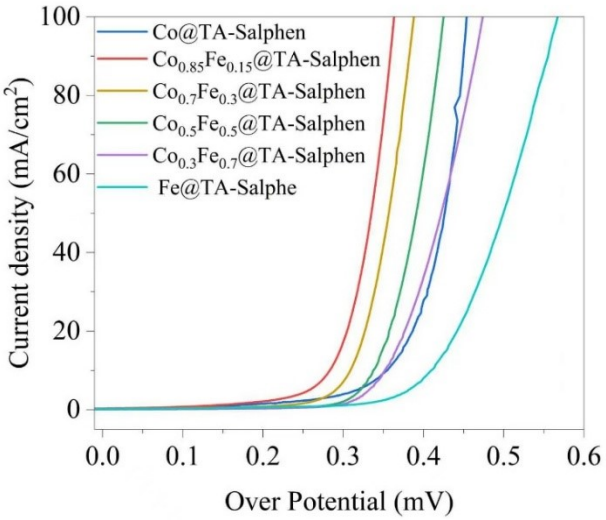

Figure S3. Comparison of OER activities for catalysts with varying Co/Fe molar ratios.

Table S1 Comparison of OER and ORR performance of $Co_{0.85}Fe_{0.15}$@TA-Salphen with other reported electrocatalysts in 1 M KOH and 0.1 M KOH.

Catalyst	Overpotential （mV）	Half-wave potential （mV）	Ref
$Co_{0.85}Fe_{0.15}$@TA-Salphen	282	923	This work
S,N-Co@CNT	276	874	1
PA-CoFe@NPC	384	850	2
Co-HAT-CN	350	800	3
Co_3O_4/CoNGDY	313.2	854	4
N-Co_3O_4@NC	266	770	5
Co_3-NG	366	903	6

FeCoCD/CSs	295	980	7
N-CNT@MOF-Co/HO-BN/CNFs	310	840	8
NC@GC	340	930	9
Ru-RuO_2/NGr	280	808	10
HESA	270	870	11
MoS_2@Fe-N-C	360	840	12
Co−C_3N_4/CNT	380	860	13
Fe Sas HS	330	860	14
$MnCo_2O_4$/NCNTs	350	760	15
Co_3HITP_2	360	800	16
Fe_2/Co_1-GNCL	350	846	17
Co_3O_4@x HoNPs@HPNCS-60	834	280	18

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