Supporting Information

Copper Complexes Covalently Grafted on Carbon Nanotubes and Reduced Graphene Oxide Promotes Oxygen Reduction Reaction Activity and Catalyst Stability

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For RDE measurement:

Koutecky–Levich plots (\mathcal{J}^1 vs. $\omega^{-1/2}$) were analyzed at various electrode potentials. The slopes of their best linear fit lines were used to calculate the number of electrons transferred (*n*) on the basis of the Koutecky-Levich equation:

$$\frac{1}{J} = \frac{1}{J_K} + \frac{1}{J_L} = \frac{1}{J_K} + \frac{1}{B\omega^{1/2}}$$

where J is the measured current density, $J_{\rm K}$ and $J_{\rm L}$ are the kinetic and diffusion-limiting current densities, $B = 0.62 nFCo_2 D^{2/3} v^{-1/6}$, ω is the angular velocity of the disk ($\omega = 2 \pi$ N, N is the linear rotation speed), n is transferred electron number, F is the Faraday constant, Co_2 is the concentration of dissolved oxygen in electrolyte, D is the diffusion coefficient of dissolved oxygen, and v is the kinematic viscosity of the electrolyte. (0.1 M KOH, $Co_2 =$ 1.2×10^{-6} mol cm⁻³, $Do_2 = 1.9 \times 10^{-5}$ cm² S⁻¹, v = 0.01 cm² S⁻¹, F = 96485 Cmol⁻¹)

For RRDE measurements:

The H_2O_2 yield and the electron transfer number (*n*) were calculated from RRDE data following the equations:

$$H_2 O_2 \% = \frac{2 \times I_r}{(N \times I_d) + I_r} \times 100$$
$$n = 4 \times \frac{I_d}{I_d + I_r / N}$$

where I_r is the ring current, I_d is the disk current, and N is the collection efficiency.



Figure S1 (A) Cyclic voltammograms of {CNTs+TAmPyCu} in Ar (dotted black) and O₂saturated (red solid) 0.1 M KOH solution. (B) Rotating-disk voltammograms of {CNTs+TAmPyCu} (loading 1.0 mg cm⁻²) in O₂-saturated 0.1 M KOH solution at different rotation rates indicated. (C) Koutecky–Levich plots at different potentials for {CNTs+TAmPyCu}. Scan rate, 10 mVs⁻¹. Theoretical $2e^{-}$ and $4e^{-}$ reduction processes are shown as dotted lines.



Figure S2 (A) Cyclic voltammograms of pristine MWCNTs in Ar (dotted black) and O₂saturated (red solid) 0.1 M KOH solution. (B) Rotating-disk voltammograms of MWCNTs (loading 1.0 mg cm⁻²) in O₂-saturated 0.1 M KOH solution at different rotation rates indicated. (C) Koutecky–Levich plots at different potentials for MWCNTs. Scan rate, 10 mVs⁻¹. Theoretical 2*e*⁻ reduction processes are shown as dotted lines.



Figure S3 (A) Cyclic voltammograms of pristine rGO in Ar (black dash) and O₂-saturated (red solid) 0.1 M KOH solution. (B) Rotating-disk voltammograms of rGO (loading 1.0 mg cm⁻²) in O₂-saturated 0.1 M KOH solution at different rotation rates indicated. (C) Koutecky–Levich plots at different potentials for rGO. Scan rate, 10 mVs⁻¹. Theoretical $2e^{-1}$ and $4e^{-1}$ reduction processes are shown as dotted lines.



Figure S4 (A) Cyclic voltammograms of {rGO+TAmPyCu} in Ar (black dash) and O₂saturated (red solid) 0.1 M KOH solution. (B) Rotating-disk voltammograms of {rGO +TAmPyCu} (loading 1.0 mg cm⁻²) in O₂-saturated 0.1 M KOH solution at different rotation rates indicated. (C) Koutecky–Levich plots at different potentials for {rGO+TAmPyCu}. Scan rate, 10 mVs⁻¹. Theoretical $2e^{-}$ and $4e^{-}$ reduction processes are shown as dotted lines.



Figure S5 Current–time chronoamperometric response of rGO-TAmPyCu (red) and 20% Pt/C (black) upon the addition of 2.0 M methanol in an O₂-saturated 0.1M KOH solution. The arrow indicates the introduction of methanol. Electrode rotation speed, 900 rpm.



Figure S6 Current–time chronoamperometric response of CNTs-TAmPyCu (black) and 20% Pt/C (red) upon the addition of 2.0 M methanol in an O₂-saturated 0.1M KOH solution. The arrow indicates the introduction of methanol. Electrode rotation speed, 900 rpm.