Supporting Information

Advances in Carbon Nitride-Based Materials and their Electrocatalytic Applications

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Table S1. Comparison of the carbon nitride-based and other types of electrocatalysts in fuel cells.

CN-based Electrocatalysts	Catalytic Performance		Reaction	Ref.
g-C_3N_4@RGO	13.7 mA cm^{-2} current density	Long-term duration stability	MOR	[1]
CN_x/PVA	6.41 mA cm^{-2} current density	High stability after 5000 repetitions	ORR	[2]
Ni/CN Cu-Ni/CN Cu/CN	58.79 A g^{-1} 5.51 A g^{-1} 1.71 A g^{-1} current densities	Stable and inexpensive	MOR	[3]
ep-GCN-AB (exfoliated porous-graphitic carbon nitride-acetylene black)	14.74 W m^{-3} power density	Improved stability	ORR	[4]
MnO_2/TiO_2/g-C_3N_4/GAC(granular activated carbon)	1680 mW m^{-3} power density	Stable after 3 weeks of operation	ORR	[5]
PtNPs@g-C_3N_4-ZIF-67 (zeolitic imidazolate framework)	375.15 mA cm^{-2} current density	Excellent long-term stability	Butanol oxidation	[6]
Ni/(g-C_3N_4/VC)	122 mA cm^{-2} current density	Stable current density	Urea oxidation	[7]
Cu_2O-g-C_3N_4/(VC)	25.3 mA cm^{-2} current density	Increased stability of about 2 h	Urea oxidation	[8]
CNQD-PANI (g-carbon nitride quantum dot - polyaniline)	28.4 A g^{-1} current density	8% loss upon 1000 cycles	MOR	[9]
C-N/CuAg/Cu_2O	1.7 mA cm^{-2} current density	Stable system	MOR	[10]
Pd-CNNF-G (carbon nitride nanoflakelets-rGO)	1890 mA mg^{-1} current density	Long-term stability	ORR	[11]

CN_x/PAN (polyacrylonitrile nanofiber)	5.82 mA cm^{-2} current density	Improved electrocatalytic stability	ORR	12
Other Electrocatalysts	**Catalytic Performance**		**Reaction**	**Ref.**
HC-LSM cathode (hybrid catalyst of $BaCe_{0.8}Gd_{0.2}O_{3-\delta}$ and $BaCO_3$ developed on $(La_{0.8}Sr_{0.2})_{0.95}MnO_{3+\delta}$ (LSM) cathodes)	Power density of 1.31 W cm^{-2} at 750 ºC	Enhanced LSM cathodes operational stability under accelerated chromium poisoning conditions	ORR	13
$Ni_{0.7}Co_{0.3}/Al_2O_3$+(10% La_2O_3) bimetallic catalyst	Power density of 651 mW cm^{-2} at 850 ºC	The presence of carbon in the anode influenced the cell long-term stability	Hydrocarbon reforming	14
Pt/Ru-SnO_2 Pd/Ru-SnO_2 (Pt and Pd deposited on nanocrystalline Ru doped SnO_2 support)	785.29 mA g^{-1} 181.71 mA g^{-1} current densities	Increased corrosion stability	EOR	15
MEAs (membrane electrode assembly) with 20, 40 and 60 wt% Pt/C + IrO_2	630, 666, 696 Mw cm^{-2} power densities	IrO_2 addition to the fuel cell prevented the degradation of the electrode	HOR	16
PtCo@AC-VC (PtCo based nanoparticles on AC-VC (activated carbon-vulcan carbon) supports)	73 mA cm^{-2} current density	Higher stability than other synthesized catalysts	MOR	17
Zirconia ALD treated Pt	Power density of 524 mWcm^{-2} at 0.4 V for bare MEA and 546 mW cm^{-2} for	ALD zirconia did not improve the catalytic performance, yet inactivated the surface as an inert surface	HER	18

	2ALD MEA after 20k cycles of accelerated stress tests	coating-Increased stability		
Pd_2-Y_2O_3/C	Power density of 325 $mWcm^{-2}$	High stability of the catalyst after 2000 operation cycles	ORR	[19]
MEA with 125 µg cm^{-2} modified MWCNT and loading of 120 µg cm^{-2} Pt/PtRu (both cathode and anode)	Power density of 1.23 W cm^{-2}	-	HOR ORR	[20]
Fe-N-MPC FeMn-N-MPC (Fe-N-mesoporous carbon)	473 mW cm^{-2} 474 mW cm^{-2}	Excellent stability after 10000 potential cycles	ORR	[21]

Table S2. Comparison of the carbon nitride-based and other types of electrocatalysts in metal-air batteries.

CN-based Electrocatalysts	Catalytic Performance		Reaction	Ref.
Pd-CN	Specific capacity of 26614 mAh g^{-1} at current density of 100 mA g^{-1}	Good cyclic stability for 70 cycles	ORR and OER	22
Fe/N doped mesoporous carbon nanosheets	Power density of 153 mW cm^{-2} Specific capacity of 628 mAh g^{-1}	Reasonable stability	ORR	23
$Ag/g\text{-}C_3N_4/Co_3O_4$	Discharge capacity of 7723 mAh g^{-1}	High cycling stability (51 cycles)	ORR and OER	24
$g\text{-}C_3N_4/\alpha\text{-}MnO_2$	Discharge capacity of 9180 mAh g^{-1}	Large cycle stability (40 cycles)	ORR and OER	25
Other Electrocatalysts	**Catalytic Performance**		**Reaction**	**Ref.**
$Pb_2[Ru_{2-x}Pb_x]O_{6.5}$ extended-pyrochlore nanocrystals on the rGO	Limiting current density of 5.1 mA cm^{-2}	Durable ORR performance-superior stability of rGO/PRO to $\alpha\text{-}MnO_2$ or Co_3O_4	ORR and OER	26
Fe/Ni(1:3)-NG)	Power density of 164.1 mW cm^{-2} Specific capacity of 824.3 mAh g^{-1}	Advantageous stability	ORR and OER	27
Ni@Co-MNC/CNTs	Current density of 10 mA cm^{-2}	Excellent rechargeability over 120000 s	ORR and OER	28
CNTs-grafted FeC/MnO_2	Power density of 35.0 mW cm^{-2}	Continuous discharge for 18 h	ORR, OER, and HER	29
$NT\text{-}Co_3O_4/NC$	Limited current density of 4.84 mA cm^{-2}	Good charging-discharging stability	ORR and OER	30

	Power density of 267 mW cm^{-2}			
$NiCo_2O_4/Mo_2C/CC$	Capacity of 778 mAh g^{-1} Power density of 104 mW cm^{-2}	Long cycle life and stability	ORR, OER, and HER	[31]
Co_3O_4/MnO_2 nanorods	116 mW cm^{-2}	Highly durable	ORR	[32]
N, S and P tri-doped graphene (NSP-Gra)	Power density up to 225 mW cm^{-2}	Long charge-discharge cycling stability	ORR and OER	[33]
NiCoFe-LDH/Ti_3C_2 MXene/NCNT	Power density of 63 mW cm^{-2} Specific capacity of 753 mAh g^{-1}	Long-term stability	ORR and OER	[34]

Table S3. Comparison of the carbon nitride-based and other types of electrocatalysts in water splitting.

CN-based Electrocatalysts	Catalytic Performance		Reaction	Ref.
$WC/Co_3W_3N/Co@NC$	10 mA cm^{-2} at only 1.61 V cell voltage	Stability for about 50 h	HER and OER	[35]
$CoS_2/g\text{-}C_3N_4$	Cell voltage of 1.6 V mV for 10 mA cm^{-2} current density	96 h of continuous electrolysis	HER and OER	[36]
MoC@NCS	1.69 V at 10 mA cm^{-2} current density	Strong durability during 24 h	HER and OER	[37]
$Cu_3P/g\text{-}C_3N_4/3D$	Cell voltage equal to 1.54 V for reaching 10 mA cm^{-2} current density	Minimum stability of 35 h	HER and OER	[38]
Other Electrocatalysts	**Catalytic performance**		**Reaction**	**Ref.**
CoP_3 nanoneedle array	Partial current density over 1 A cm^{-2}	Excellent stability	HER and OER	[39]
RuNiCo@CMT	Overall cell voltage of 1.58 V at 10 mA cm^{-2}	Excellent stability for up to 30 h	HER and OER	[40]
Co-Fe Selenide	Cell voltage of 1.68 V at a current density of 10 mA cm^{-2}	Stability for 9 h	HER and OER	[41]
$CuS/MnCO_3$ on nickel foam	10 mA cm^{-2} at a voltage of 1.43 V	Stability for about 10 h	HER and OER	[42]
FeCoMoS@NG	Cell voltage of 1.58 V at current density of 10 mA cm^{-2}	Durability over 70 h of successive charge-discharge cycles	HER and OER	[43]
$Fe\text{-}N\text{-}C/FeP_x/NPSC$	1.57 V cell voltage to reach 10 mA cm^{-2}	High reliability under various deformation conditions	ORR, HER and OER	[44]
$CC/MOF\text{-}CoSe_2@MoSe_2$	Voltages of 1.53 and 1.76 V to accomplish current	Excellent stability over 24 h for OER, HER,	HER and OER	[45]

	densities of 10 and 50 mA cm^{-2}	and overall water splitting		
Pt-WO_{3-x}@rGO	Working voltage of 1.55 V at 10 mA cm^{-2}	Durability over 12 h of continuous operation	HER and OER	46
$FeWO_4$-Ni_3S_2	1.51 V at 10 mA cm^{-2}	Operating stably for 100 h at 1000 mA.cm^{-2}	HER and OER	47
$CuMo_2ON$@NG	Cell voltage of 1.49 V at a current density of 10 mA cm^{-2}	Excellent reversibility of 120 h at a high current density of 100 mA cm^{-2}	ORR, HER and OER	48

Table S4. Comparison of the carbon nitride-based and other types of electrocatalysts in a number of oxidation/reduction reactions.

CN-based Electrocatalysts	Catalytic Performance		Reaction	Ref.
Fe_3C@mCN	Current density of -3 mA cm^{-2}	90% current retention after10 h	ORR	[49]
Fe/Ni@g-C_3N_4	Current density of 3.6 mA cm^{-2}	Strong mechanical stability	ORR	[50]
Pd-g-C_3N_4/NCQD	Mass activity of 600 A g^{-1}	Near 10.5% loss after 10 h reaction	ORR	[51]
Fe-N-C	Power density of 0.086 Wcm^{-2} at 0.18 mA cm^{-2} as fuel cell	1.3% performance decrease after 2000 cycles	ORR	[52]
Co_2@CN	Limiting potential of -0.78 V	High stability and efficient N_2 conversion	NRR	[53]
Boron carbon nitride (BCN)	-9.87% Faradaic efficiency -41.9 µg $h^{-1}mg_{cat}^{-1}$ ammonia yield	Noticeable stability	NRR	[54]
Mn-C_3N_4/CNT	98.8% Faradic efficiency and 14 mA cm^{-2} current density at 0.44 V	After 20 h no obvious decay in current density	CO_2RR	[55]
Ag-S-C_3N_4/CNT	Current density of -21.3 mA cm^{-2} at -0.77 V Faradaic efficiency of 90%	No significant decay after 24 h	CO_2RR	[56]
Other Electrocatalysts	**Catalytic Performance**		**Reaction**	**Ref.**
$Ni(OH)_2/ZrO_2$	Current density of -3.88 mA cm^{-2}	Excellent stability and methanol tolerance under alkaline conditions	ORR	[57]
Si-Fe-N/C	Current density of 5.4 mA cm^{-2}	Excellent durability (90% after 50000 s)	ORR	[58]

Fe-N-WC	Current density of 6.644 mA cm^{-2}	Maintained 96.5% of the initial current after 10000 s	ORR	59
Ru-Co_3O_4	Current density of 3 mA cm^{-2}	86.0% for 20 h	ORR	60
TiO_2/rGO	Faradic efficiency of 8.88% NH_3 yield of 7.75 µg $h^{-1}.cm^{-2}$	Less than 10% drop after six recycling tests	NRR	61
Zr/α-FeOOH	NH_3 yield of 1.39×10^{-10} mol $s^{-1}cm^{-2}$ Faradic efficiency of 35.63%	No significant change after 5 cycles	NRR	62
Cu/CeO_2	49.3% Faradaic efficiency at -1.6 V (current density of about 12 mA cm^{-2})	Relatively stable in 210 min	CO_2RR	63
Cu_2O/graphene	Faradaic efficiency of 93.20% at -1.0 V	Stable current after 8 h	CO_2RR	64

Table S5. Comparison of the carbon nitride-based and other types of electrocatalysts in supercapacitors.

CN-based Electrocatalysts	Catalytic Performance		Ref.
g-C_3N_4/PPy	Areal capacity of 289.6 mF cm^{-2} at current density of 0.4 mA cm^{-2}	Life cycles after 10000 cycles	65
g-C_3N_4@$ZnCo_2O_4$	Specific capacity of 157 mAh g^{-1} at a current density of 4 A g^{-1} Using Symmetric device: 39 Wh kg^{-1}energy density and 1478 W kg^{-1} power density	After 2500 cycles about 90% capacity retention and 71% capacity retention after 10000 cycles	66
Other Electrocatalysts	**Catalytic Performance**		**Ref.**
Zn//PBC-A900	Super capacitance of 321.3 F g^{-1} at 1 A g^{-1} Energy density of 114.2 Wh kg^{-1} with the power density of 800 W kg^{-1}	78% capacitance retention after 20,000 cycles	67
CFt/PPy/ASA	Specific areal capacitance of 4000 mF cm^{-2} Energy density of 18.8 Wh kg^{-1} at a power density of 1875 W kg^{-1}	Capacitance retention of 82.7% after 2500 cycles	68
Copper wire@CuO@MnO_2	Capacitive performance of 5.97 F cm^{-3} Energy density of 0.38 mWh cm^{-3} at a power density of 25.5 mW cm^{-3}	Cycling stability of 77% after 2000 cycles	69
PAN/PMMA/Ultra-Fine Needle Coke	Specific capacity of 387.2 F g^{-1} at 0.5 A g^{-1} Energy density of 27.87 Wh kg^{-1} at 489 W kg^{-1}	97.5% capacity retention after 10,000 cycles at 1 A g^{-1}	70
N-PCMs	158 F g^{-1} under the condition of 0.5 A g^{-1}	Capacitance retention rate of 89.4% after 10000 cycles at 1 A g^{-1}	71

Table S6. List of various energy devices designed for evaluating carbon nitride-based materials for electrocatalytic reactions.

Material	Energy Device	Reaction	Electrocatalytic performance summary	Ref.
Graphene-based carbon nitride; G-CN	Fuel cell	ORR	Current density of 7.3 mA cm^{-2} at 0.40 V	72
Pt-Fe carbon nitride; $K_{0.12}[Pt_1Fe_{1.6}C_{55}N_{0.12}]$	Fuel cell	ORR HOR	Specific activity per platinum unit mass of -255 A g^{-1} Pt (0.75 V) 967 A g^{-1} Pt (<0.2 V) Activation potential of 15 mV higher than reference EC-20	73
Bimetallic carbon nitride; Pt-Ni-CN and Pt-Fe-CN	Fuel cell	ORR	Low overpotential (~40 mV), high open circuit potential and mass activity at 0.9 V, low Pt mass for power production (e.g.0.099 A mg_{Pt}^{-1} and 0.86 g_{Pt} kW^{-1} for Pt-Fe-CN 900)	74
Pt-Fe and Pt-Ni carbon nitride-based core-shell; PtNi-CN and $PtFe_2$-CN	Fuel cell	ORR	Lowest overpotential by PtNi-CN_l 900/Graphene (30 mV lower than Pt/C), Low Pt mass for power production (0.3 g_{Pt} kW^{-1} for PtNi-CN_l 900/G, Higher activation potential (30 mV) than reference MEA	75
Pt/carbon nitride (CN_x) modified SiO_2; CN_x/SiO_2	Fuel cell	ORR	2.9 and 3.21 times higher mass activity by Pt/CNx/SiO_2 than Pt/C at 0.85 V and 0.8 V, respectively	76
Chain-like SnO_2-CN_x; SnO_2-CN_x	Fuel cell	ORR	1.6 and 1.2 times larger specific activity than Pt/C and Pt/C-CNx, respectively	77
Hollow mesoporous carbon nitride nanosphere/graphene; HMCN-G	Fuel cell	ORR	Positive onset potentials of -0.15 to -0.1 V and high current density up to -3.3 mA cm^{-2} at -0.6 V	78

Coupled Co-graphitic-carbon nitride carbon nanotubes; Co-g-C_3N_4/SWCNTs	Fuel cell	ORR	More positive onset potential (-0.03 V) and half-wave potential (-0.15 V) than Pt/C	[79]
Pd nanoparticles-graphitic-carbon nitride; PdNPs/CNx	Fuel cell	FAO	High mass activity of 1640 mA mg_{Pd}^{-1} (7.2 times higher than Pd/C) with a peak potential of 70 mV (23 times higher mass activity than Pd/C after 1000 cycles)	[80]
Nano-electrocatalysts on carbon nitride support; PtNi-CN/G	Fuel cell	ORR	About 30 mV lower overpotential than Pt-based electrodes, 0.3-0.4 g of Pd or Pt for 1 kW production by PtNi-CN900/G	[81]
Bimetallic core-shell carbon nitride; PtNi-CN/G and $PtFe_2$-CN/G	Fuel cell	ORR	High selectivity/high neighboring Pt sites pairs density by PtNi-CN600/G and $PtFe_2$-CN600/G	[82]
Graphene supported Co-g-C_3N_4; CCNG-600	Fuel cell	ORR	Onset potential of -0.03 V and half-wave potential of -0.141 V	[83]
Nitrogen-doped graphene supported graphitic carbon nitride; g-C_3N_4/N-G	Fuel cell	ORR	Onset potential of -0.02V and half-wave potential of -0.22V, high current retention (96.3%) even after 30000 s	[84]
Co_3O_4 and carbon dot nanocrystals on graphitic carbon nitride; Co_3O_4-C/C_3N_4	Fuel cell	ORR	Electron-transferred number of 3.69 at -0.8 V and onset potential of -0.09V, large current density	[85]
RuO_2 nanowires on graphitic carbon nitride; 1D-RuO_2-CN_x	Water splitting	OER	Low onset overpotential of 200 mV with Tafel slopes of 52 and 56 mV dec^{-1} and low overpotentials of 250 mV and 260 mV for 10 mA cm^{-2} current density in acidic and basic media, respectively, Mass activity of 352	[86]

		HER	mA mg^{-1} (about 14 times higher than RuO_2) Superior HER activity with current density of 10 mA cm^{-2} at 93 and 95 mV and Tafel slopes of 40 mV dec^{-1} and 70 mV dec^{-1} in acidic and basic condition, respectively	
Cu-doped carbon nitride; Cu-g-C_3N_4	Water splitting	HER	High current density of 10 mA cm^{-2} at a low overpotential of 390 mV with good performance in acidic media for at least 43 h	87
Cobalt/nitrogen-doped graphitic carbon; Co-NG	Water splitting	HER	Overpotential of 203 mV at 10 mA cm^{-2} current density 2.5 times PHE rate by Co-NG/g-C_3N_4 than pure g-C_3N_4	88
Coral-like-nickel-carbon nitride; Ni/C_3N_4	Water splitting	HER	High exchange current density of 1.91×10^{-4} A cm^{-2}, low Tafel slope of 128 mV dec^{-1}, and low overpotentials of 356 and 222 mV for 100 and 10 mA cm^{-2} current densities	89
Bimetallic nanoparticles decorated carbon nitride; C_3N_4/AgPt, C_3N_4/AgPd, and C_3N_4/AgAu	Water splitting	HER	Low overpotential of -150 mV to attain 10 mA cm^{-2} current density by C_3N_4/AgPt with Tafel slope of 65 mV dec^{-1}, Activity order: 5%Pt/C>C_3N_4/AgPt>C_3N_4/AgPd> C_3N_4/AgAu	90
Pd and Pt nanoparticles decorated carbon nitride; C_3N_4/Pt and C_3N_4/Pd	Water splitting	HER	-0.339 V and -0.371 V potentials for 10 mA cm^{-2} current density by C_3N_4/Pt and C_3N_4/Pd	91

Nitrogen-doped porous carbon nanosheets templated from graphitic carbon nitride; N-CNS	Fuel cell/Battery	ORR	Superior ORR performance than Pt/C by N-CNS-120 with 11.6% nitrogen content	92
Hierarchically porous graphene sheets-graphitic carbon nitride; hp-GS/GCN	Fuel cell/Battery	ORR	Low half-wave potential and highest potential when current density is higher than Pt/C	93
Fe-carbon nitride Core-shell; $FeFe_2$-CN/CA	Fuel cell/Battery	ORR	Onset potential of 0.908 V by $FeFe_2$-CN900/CA (38 mV lower than Pt/C)	94
Boron-doped graphitic-carbon nitride; B_x-GCN	-	HER	Reduction of *p*-nitrophenol with high electrocatalytic activity and stability	95
TiO_2 nanotube/graphitic-carbon nitride	Battery	HER	Superior PEC activity towards Rhodamine B dye reduction	96
Graphitic-carbon nitride/carbon paper; GCN/CP	Li-O_2 Battery	ORR	Decreased voltage gap of 0.95 V as discharged to 1000 mAh g^{-1} at 200 mA g^{-1} current density and 1.58 V at 2000 mA g^{-1}	97
Carbon felt modified with carbon nitride; C_3N_4-CF	RF Battery	VO_2^+/VO^{2+} redox	Ipa/Ipc about 0.93 with the polarization of 0.16 V, energy efficiency (EE) of 85% by C_3N_4-CF (9.1% higher than pure CF)	98
Mesoporous graphitic-carbon nitride/NiS quantum dot; g-C_3N_4/NiS	Solar cell	Sn^{2-} reduction	Low interface charge transfer resistance (Rct) of 1.08 Ω, high power conversion efficiency (PCE) of 5.64%	99

MEA: Membrane electrode assemblies; FAO: Formic acid oxidation; PHE: Photocatalytic hydrogen evolution.

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