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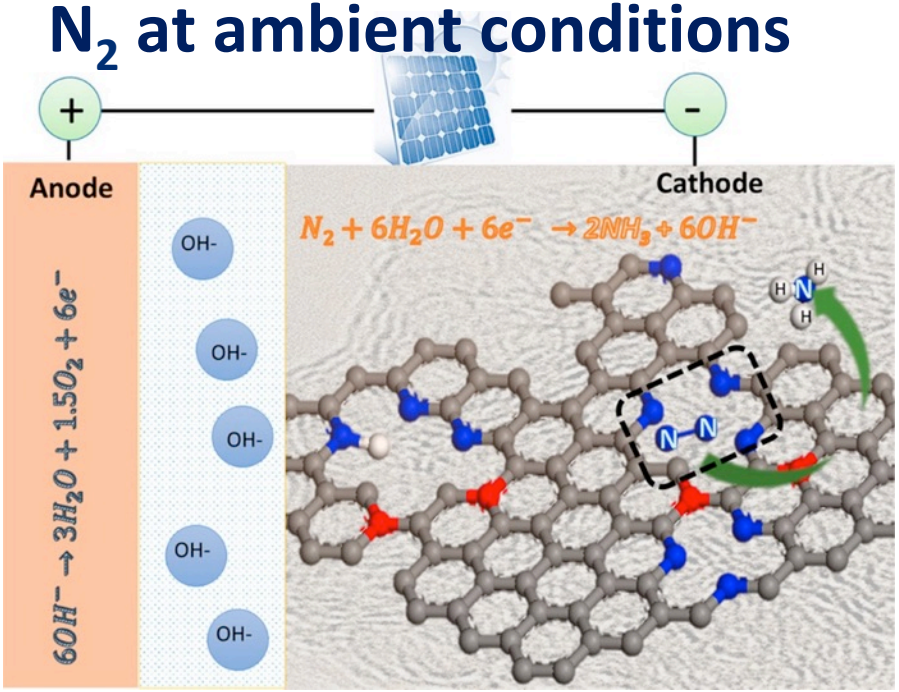
Department of Chemical and Biological Engineering
School of Engineering and Applied Sciences

Developing Electrocatalyst for NH_3 Production from H_2O and N_2 at ambient conditions



Shreya Mukherjee (PHD Candidate)
Advisor: Gang Wu

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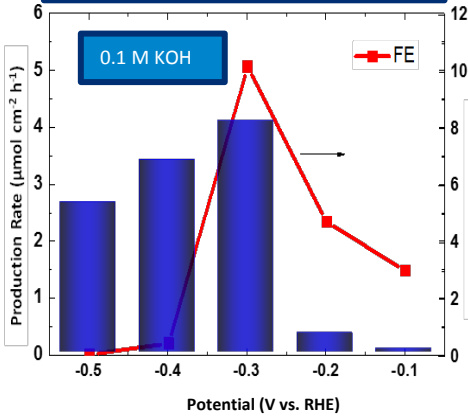
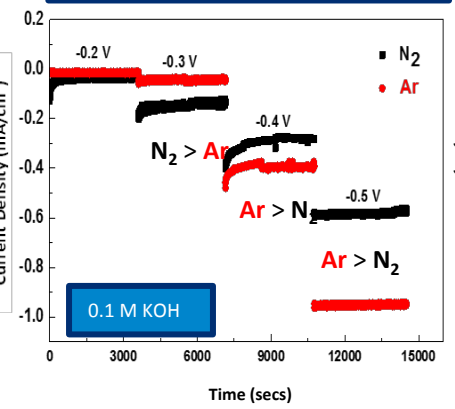
Outline of Development of NRR catalysts

Necessity Of Determining Optimum Potential And Best Electrolyte

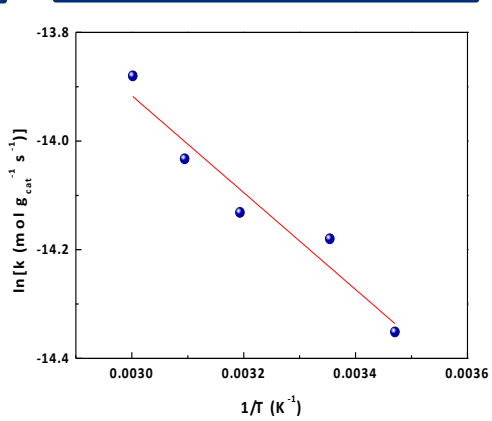
Activity at different potential



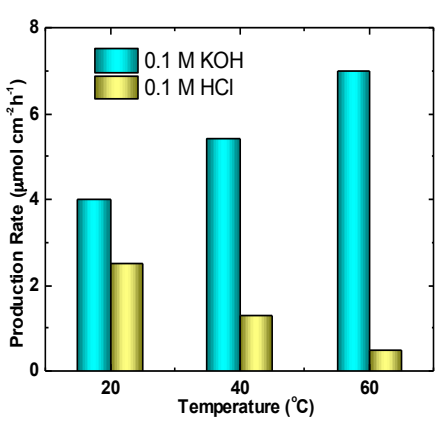
Current density with potential



Activation Energy-7.42 kJ/mol



Electrolyte: Alkali *vs.* Acid



- Controls-Tested electrolyte before testing in N_2 and Ar to consider ammonia that might be present in atmosphere
- Separate electrochemical cell for testing in N_2 and Ar
- Method of detection-Indophenol test (Also retested with salicylate method)

- Best electrolyte for our catalyst-0.1 M KOH
- Optimum Potential → (-) 0.3 V *vs.* RHE

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Outline of Catalyst Development Approach

Initial Motivation:

Metal nitrides based on DFT

Main challenge:

Catalyst decomposition

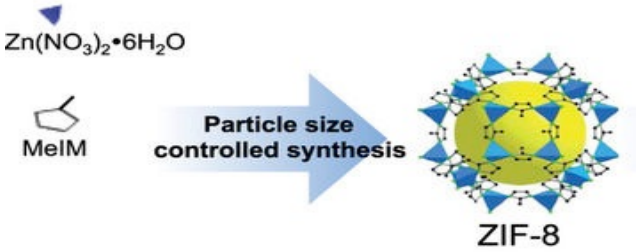
Further Modification

FeN_4 supported on stable N doped carbon derived from ZIF8

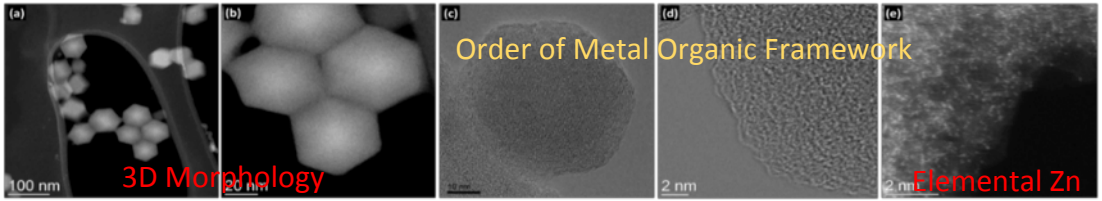
Main Challenge: Hydrogen Evolution

Innovation:

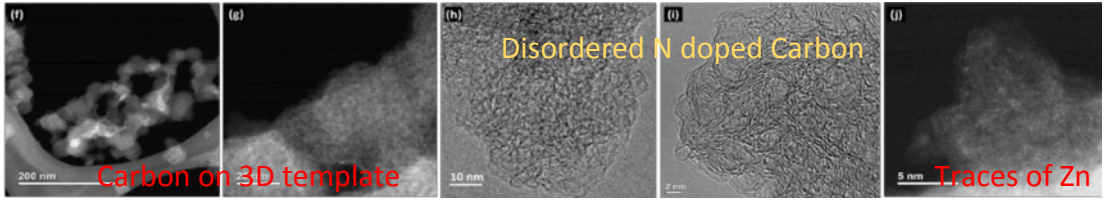
Derived carbon from Fe free ZIF8 by thermal activation



Highly disordered carbon derived from ZIF-8 via pyrolysis under inert atmosphere



ZIF-8 before pyrolysis



ZIF-8 after pyrolysis

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Outline of Catalyst Development Approach

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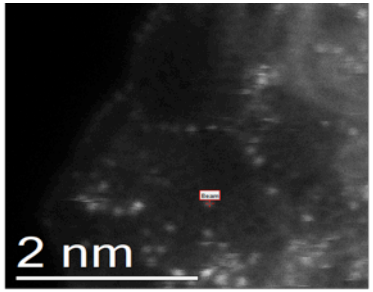
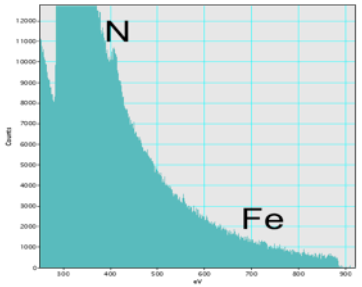
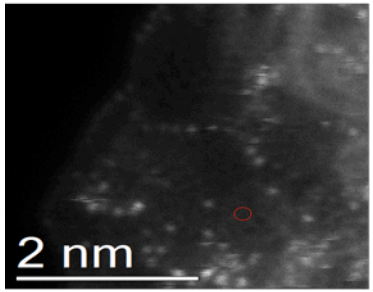
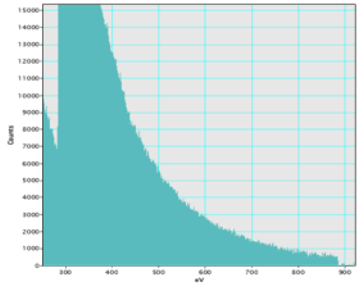
Single Atomic Iron Catalysts for Oxygen Reduction in Acidic Media: Particle Size Control and Thermal Activation

Hanguang Zhang[†], Sooyeon Hwang[‡], Maoyu Wang[§], Zhenxing Feng[§], Stavros Karakalos[⊥], Langli Luo[∥], Zhi Qiao[†], Xiaohong Xie[∥], Chongmin Wang[∥], Dong Su[‡], Yuyan Shao*[,†], and Gang Wu*[,†]
[†] Department of Chemical and Biological Engineering, University at Buffalo, The State University of New York, Buffalo,









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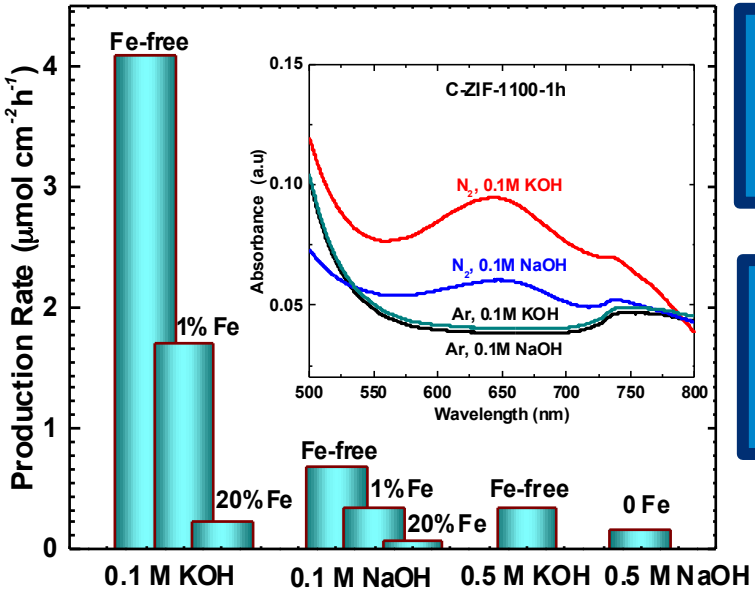
Investigated Fe-N_4 Sites on N Doped Carbon For NRR Activity



Mukherjee et al. *Nano Energy 48 (2018) 217–226*

Carbon derived ZIF8 at optimized pyrolysis condition, had highest activity

Potassium ions in electrolyte played a role in reducing hydrogen evolution

Activity increased as Fe doping was reduced

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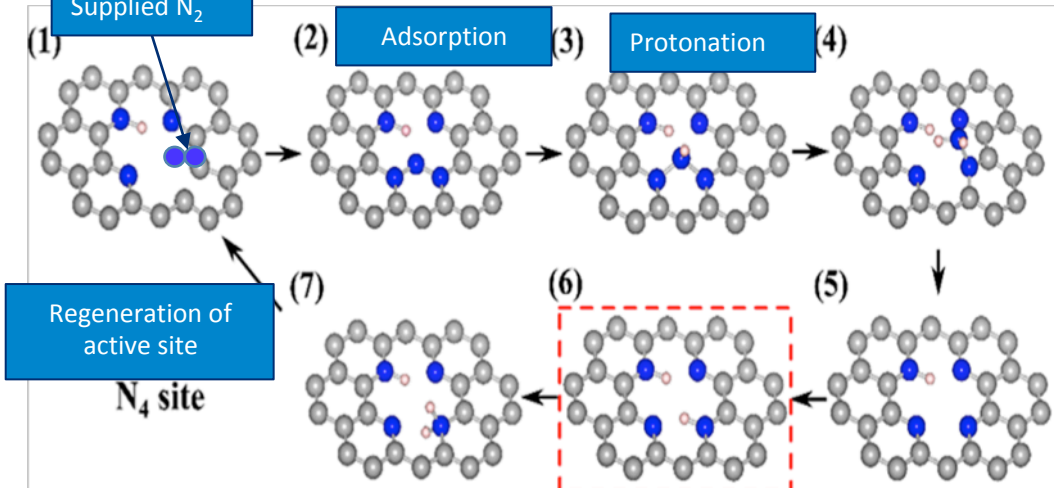
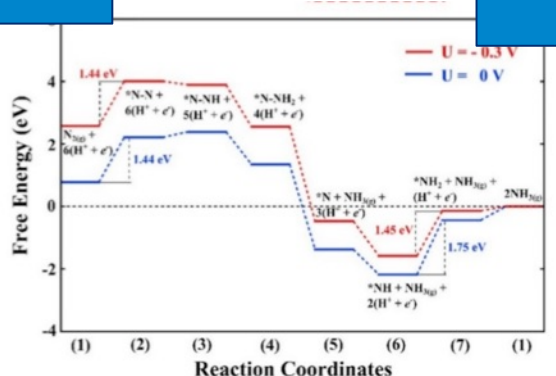
Associative Distal Pathway: Mechanism Of NRR On Carbon with Nitrogen Vacancy





(1) Pyrolic nitrogen vacancy and carbon defects originated due to pyrolysis

(2) Adsorption of N_2 on vacant site

(3) Protonation

(4) Further protonation

(5) Detachment of 1st ammonia molecule

(6) Protonation for release of second ammonia molecule is favored with applied potential

(7) Release of Ammonia regenerates the N vacancy

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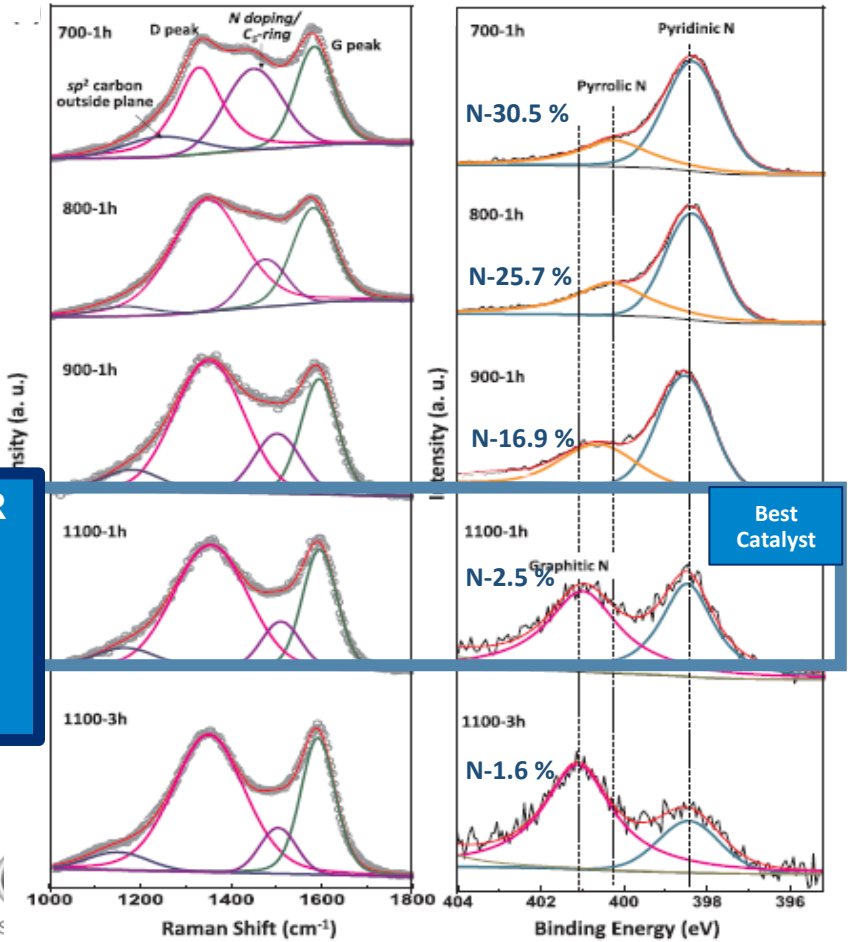
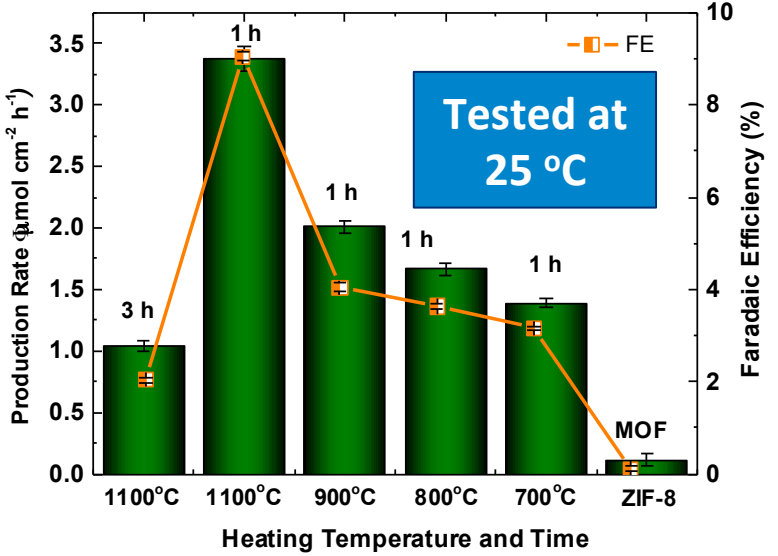
Effect Of Pyrolysis Condition on Activity



Best Catalyst



Highest NRR activity- 3.4 x10 $^{-6}$ mol $cm^{-2}h^{-1}$

F.E.~10 %

- Zn boils at 907 °C leaving more defects in the carbon above 900 °C
- Reduction of nitrogen doping with increase in pyrolysis temperature
- Removal of pyrolic nitrogen may create adsorption site for nitrogen

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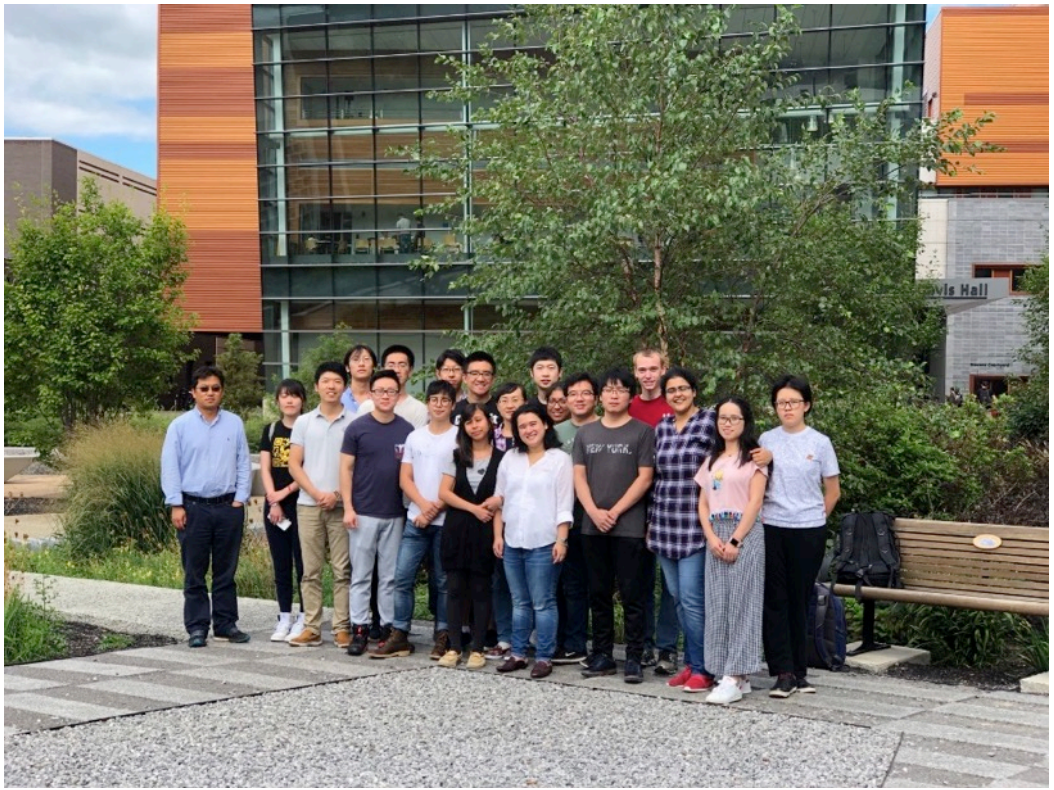
Acknowledgement



My Parents

Adviser- Dr. Gang Wu

Entire Lab Group

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