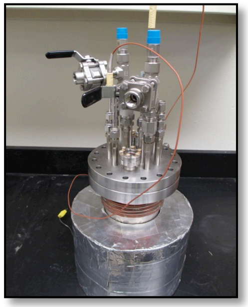

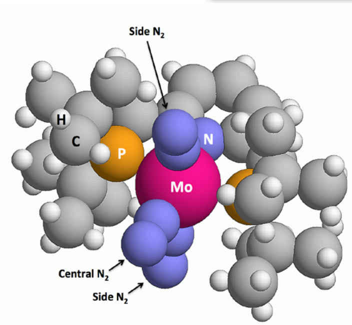

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Nitrogen/Oxygen Battery

A Transformational Architecture for Large Scale Energy Storage

US DOE Energy Storage Systems Research Program

Peer Review, Washington, DC Sept. 26-28, 2012

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NNSA National Nuclear Security Administration

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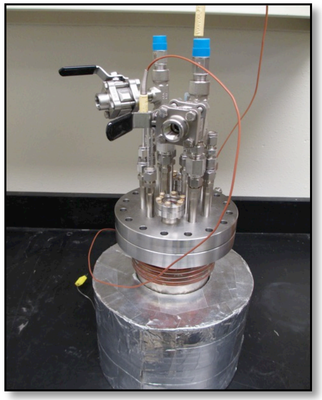
N_2/O_2 Battery Project Overview

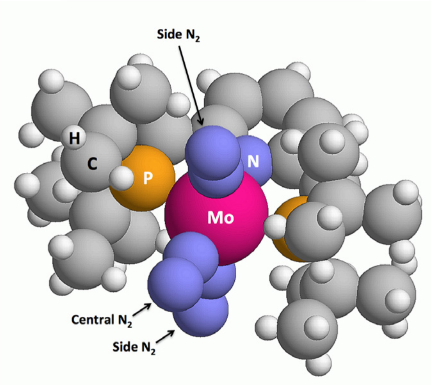
- Air/Air battery.
- N_2 electrochemistry enables the redefinition of a gas (diffusion) electrode and the three phase interface.
- Operated as redox flow battery.
- Provide a very high energy density, very low cost, environmentally benign electrochemical platform for load leveling and for grid-integrated storage of energy generated by wind, solar and other sustainable but intermittent sources.
- Project requires a reversible N_2 electrode.

Anode requires the reversible electrochemical reduction/oxidation of N_2.

Three approaches to achieve N_2 reduction (nitrogen fixation).



1) Direct reversible reduction/oxidation of N_2 to $2N^{-3}$ in molten salt at + 400°C.



2) Mediated and catalyzed reduction of N_2 to NH_3 and subsequent utilization of the NH_3 as the anode in an NH_3/air fuel cell.

3) Indirect reversible reduction/oxidation of N_2 to $2N^{-3}$ at ambient temperature.

Indirect Reduction of N_2 at Ambient Temperature.

At ambient temperature N_2 is not directly reduced on any electrode in any electrolyte.

Some researchers have achieved Li mediated reduction of N_2 by the exposure of N_2 to electrodeposited Li metal.

$$Li^+ + e^- \rightarrow \underline{Li}$$

$$3\underline{Li} + 1/2N_2 \rightarrow Li_3N \qquad \text{electrode passivates}$$

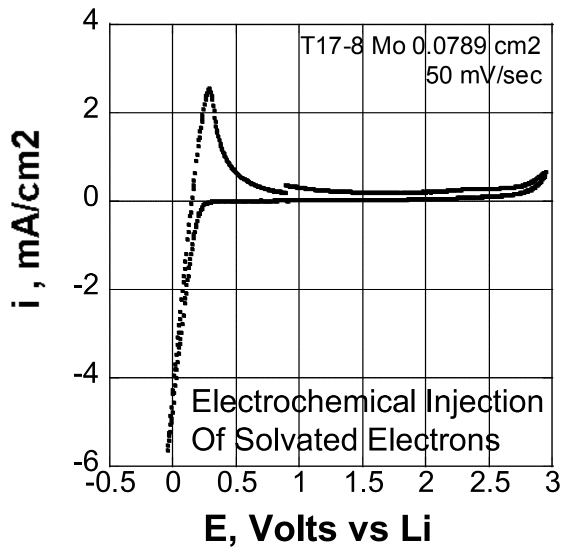
$$Li_3N + 3EtOH \rightarrow NH_3 + 3LiOEt \qquad \text{low efficiency}$$

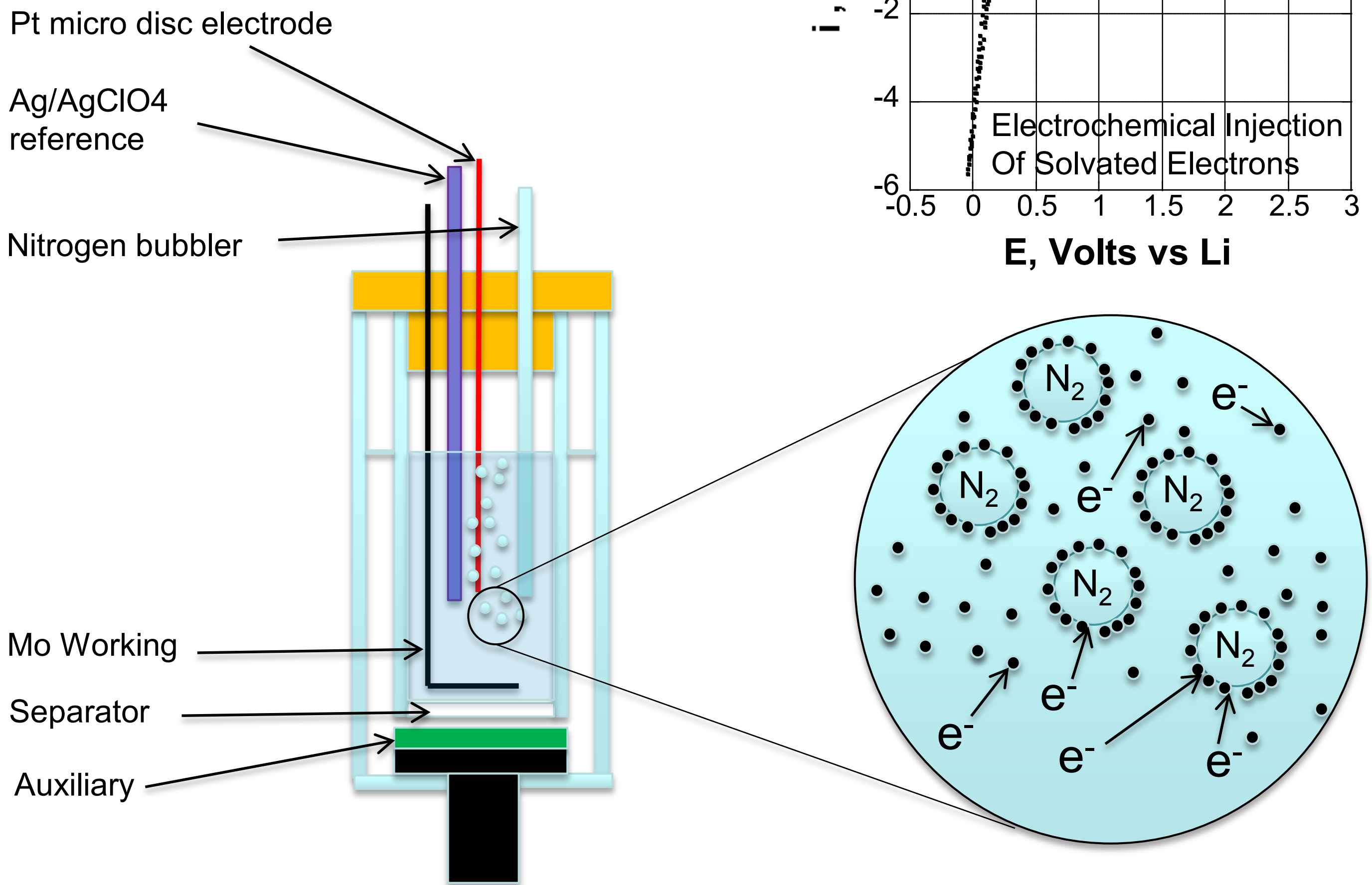
Our approach: Reduce N_2 in a solution of solvated electrons [e_s^-].

$$N_2 + 6e_s^- \xrightarrow{Li^+} 2Li_3N$$

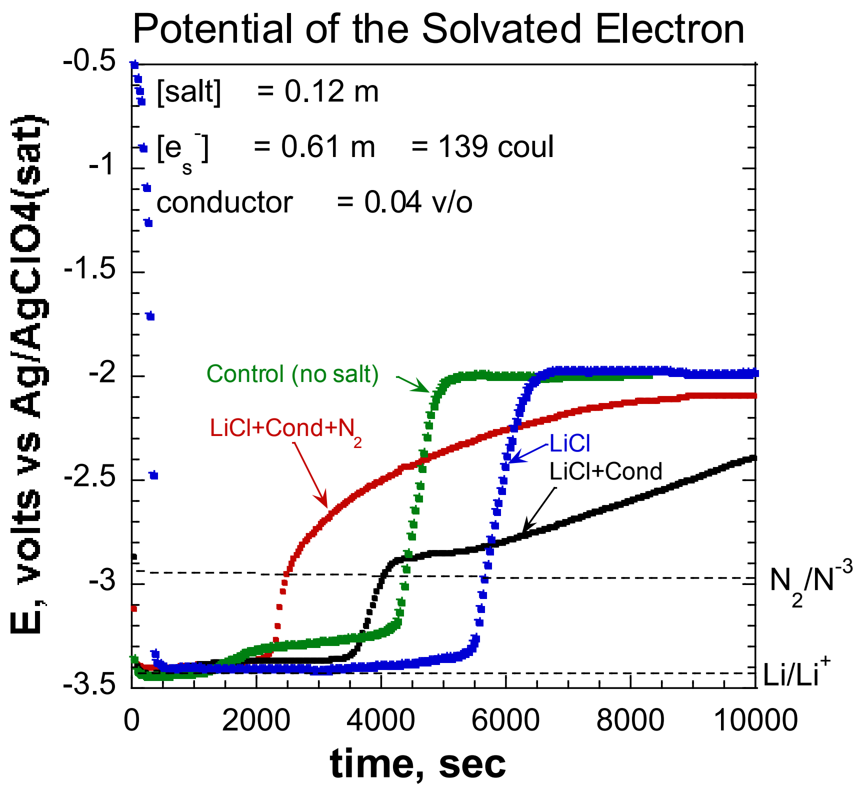
Reduction does not occur at the electrode surface.
Advantage: Gas diffusion electrode not required.

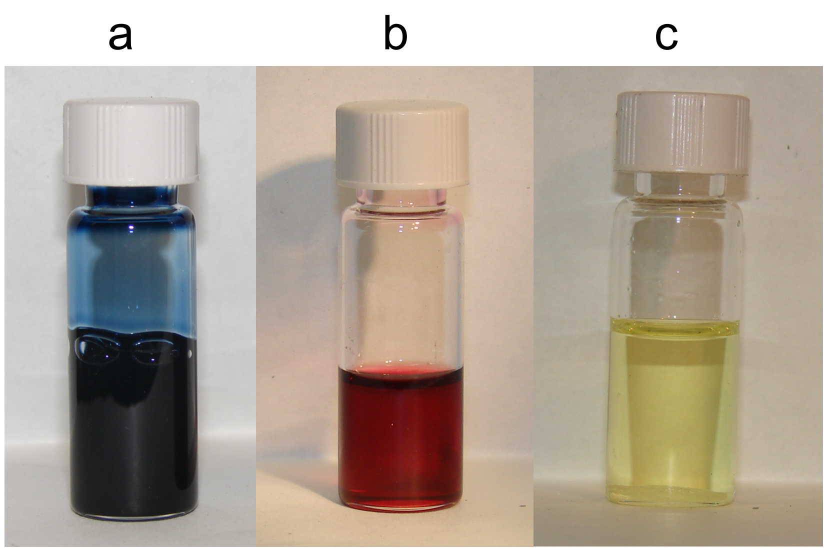
Indirect Reduction of N_2.
Concept Illustration





Stabilization of the Solvated Electron [e_s^-] at High Negative Potential Enables the Reduction of N_2





-3.4 V　-2.0 V　c = b+EtOH

Summary/Conclusions

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1) Demonstrated reversible redox for N_2/N^{-3} in LiCl-KCl at 450 C

   Developed procedure for large scale purification of LiCl-KCl eutectic.

2) Density Functional Theory (DFT) has been used to analyze the path to NH_3 synthesis on the Arashiba *et. al.* catalyst.

   Paper submitted for publication (Peter Feibelman, J. Phys. Chem.).

3) Developed an electrolyte which stabilizes solvated electrons for the subsequent indirect reduction of N_2.

Future Tasks

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- Couple N_2/N^{-3} and O_2/O_2^- reactions in LiCl-KCl electrolytes to produce N_2/O_2 battery prototypes.
- Continue stabilization of solvated electrons at high negative. potential and introduce catalysts to enhance N_2 reduction.
- Identify procedures for chemical analysis of N_2 reduction products.
- Identify appropriate cation to enable the oxidation of N^{-3}.

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Synthesis and characterization of Mo catalysts and ionic liquids.
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Harry Pratt   Advanced Power Sources Dept., SNL

Discussions on electrochemical stability of electrolytes and solvated electrons.
Dr. Chengdu Liang   ORNL
Dr. Sheng Dai   ORNL
Dr. Loic Baggetto   ORNL