Contract No:

This document was prepared in conjunction with work accomplished under Contract No. 89303321CEM000080 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

Disclaimer:

Summary Statement:
In order to lessen societies dependance on fossil fuels to fight climate change, new energy storage and electrification technology is needed. This work highlights the development of non-conventional electrolytes for lithium-ion and redox flow batteries for electric vehicle and grid storage applications.

Introduction:

The primary technology currently used for energy storage is lithium-ion batteries. Conventional rechargeable lithium-ion batteries are ubiquitous in daily life, being the battery of choice for phones, laptops, and, more recently, electric vehicles (EVs). However, lithium-ion battery research is primarily focused on using conventional organic solvents as the supporting electrolyte such as dimethyl carbonate and ethylene carbonate. Carbonate based electrolytes have numerous downsides such as safety and stability.

This project focused on the development of non-conventional electrolytes to address this shortfall. So far, this work has developed a novel deep eutectic electrolyte (DEEs) for potential use in lithium-ion batteries. The developed electrolyte has shown satisfactory performance while allowing for new pathways on the development of novel non-conventional lithium systems, and systems with potential low temperature operation. The understanding gained from this research can also be applied to non-lithium-based systems such as sodium, potassium, aluminum, magnesium, and calcium.

Building upon the knowledge gained in this research, the study of electroactive DEEs for energy grid storage applications in the form of redox flow batteries is also possible. Redox flow batteries, as compared to a standard battery, use redox active electrolytes as the catholyte and anolyte in the battery. Electroactive DEEs could offer an interesting alternative to the industry standard acidic-aqueous based vanadium electrolytes. Compared to aqueous systems, DEEs can have higher concentrations (also yielding higher capacities), less corrosive environments, and have a large electrochemical stability window.

Approach:

The approach used in this project highlights our capabilities in non-conventional eutectic electrolytes research. Specifically, it allows for the study of molecular interactions between hydrogen bond donors (in this case methyl carbamate) and acceptors with lithium salts, and their ability to form a eutectic. The formulated eutectic is then studied using various electrochemical and spectroscopic characterization techniques to identify positives and shortfalls of the system. This approach will allow the leveraging of our growing capabilities in lithium-ion batteries technologies to other non-lithium-based systems and allow SRNL to develop in the field on non-conventional electrolytes. Various electrochemical measurements such as cyclic voltammetry (CVs) and galvanostatic cycling provide us a tool to study the properties of the eutectic electrolytes. Furthermore, Impedance data will allow us to directly measure how the eutectic conductance changes with regards to cycling and

Figure 1: Coin cell setup used for electrochemical tests

temperature. Much of this work will be completed using electrochemical coin cell (Figure 1)

Accomplishments:

- Designed a novel DEEs for lithium-ion batteries based on methyl carbamate and lithium. hexafluorophosphate ($LiPF_6$) and lithium bis(trifluoromethanesulfonyl)imide (LiTFSI).
- Achieved 93% capacity retention after 50 cycles with a voltage stability between ~1 V to 5 V.
- Lithium battery was able to power LED at less than -45 °C.
- Developed eutectics based on Iron, Chromium, and Manganese salts for redox flow batteries.
- Redox flow battery eutectics showed to be electroactive using CVs.

Peer-reviewed Publications:
N. Z. Hardin, Z. Duca, A. Imel, P. A. Ward, Methyl Carbamate-Lithium Salt Deep Eutectic Electrolyte for Lithium-Ion Batteries, *ChemElectroChem* 2022, 9, e202200628.

Intellectual Property:
N/A

Total Number of Post-Doctoral Researchers:
Nathaniel Hardin, SRNL

Total Number of Student Researchers:
N/A

Figures:

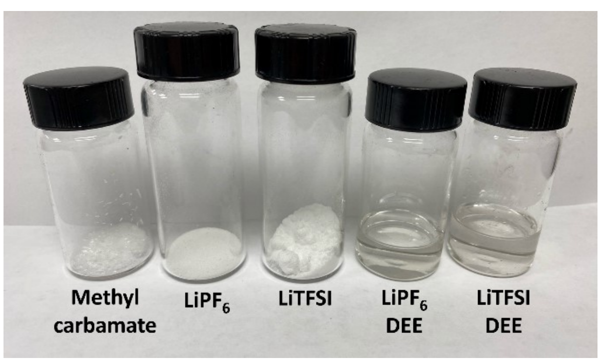

Figure 2: Picture of starting materials and formed DEEs

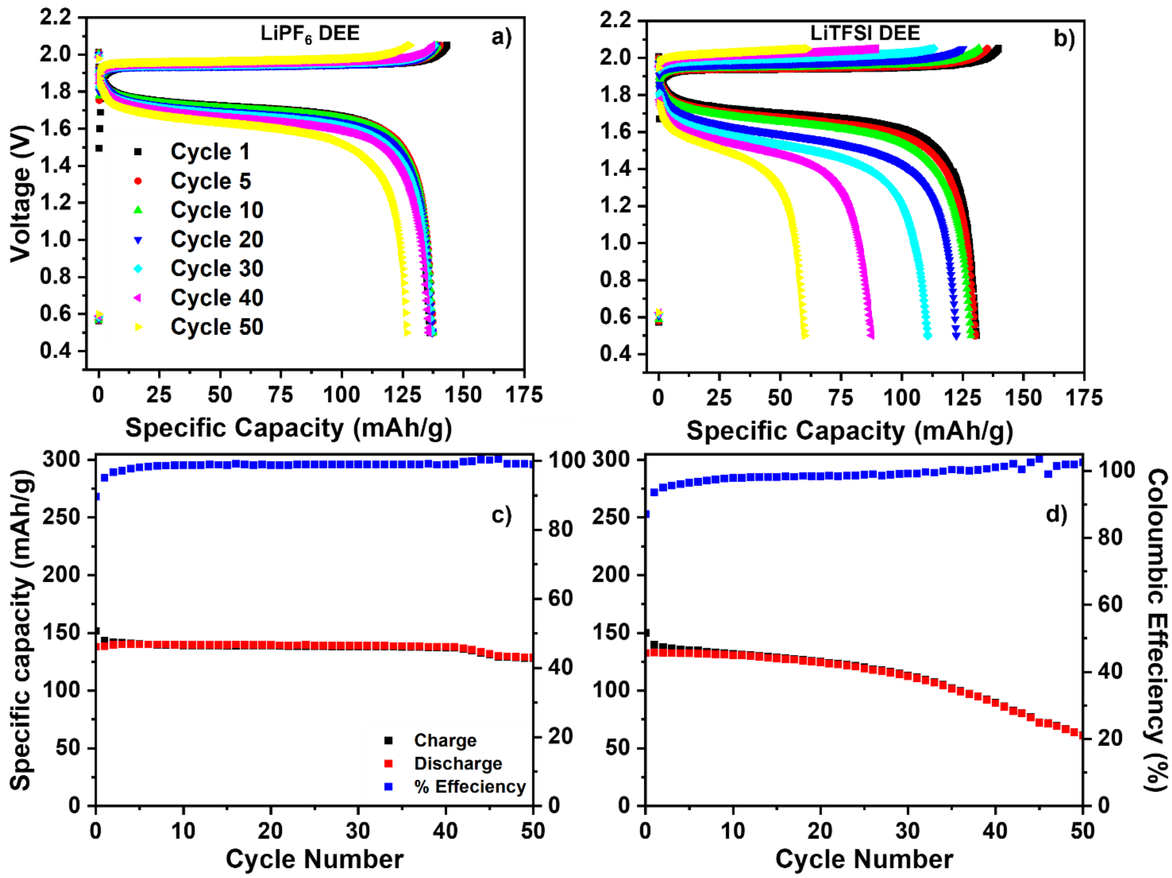

Figure 3: a-b) Voltage profiles of (a) $LiPF_6$ DEE and (b) LiTFSI DEE, c-d) Cycling performance and efficiency of (c) $LiPF_6$ DEE and (d) LiTFSI DEE.

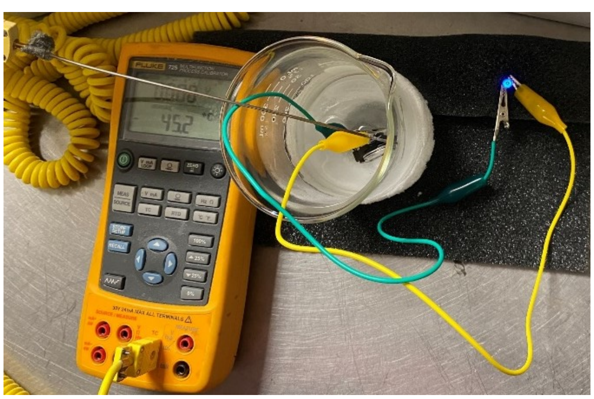

Figure 4: Low temperature operation of a blue LED

REVIEWS AND APPROVALS

1. Principal Investigator:

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Name and Signature Date

2. Technical Review:

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Name and Signature Date

3. PI's Manager Signature:

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Name and Signature Date

4. PI's Division Director Signature:

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Name and Signature Date

5. Intellectual Property Review:

This report has been reviewed by SRNL Legal Counsel for intellectual property considerations and is approved to be publicly published in its current form.

SRNL Legal Signature

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Name and Signature Date