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Study Non-Conventional Deep Eutectic Electrolytes for Lithium Ion Batteries

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Main Findings

Formation of novel deep eutectic electrolyte

To prepare the novel eutectic electrolyte based on methyl carbamate and lithium salts (LiPF₆ or LiTFSI) the starting materials were mixed together at a 1:5 molar ratio (Li(PF₆ or TFSI) : Methyl carbamate at room temperature. The two solids once mixed will spontaneously form a eutectic solution as demonstrated by the melting of the solids into one electrolyte.

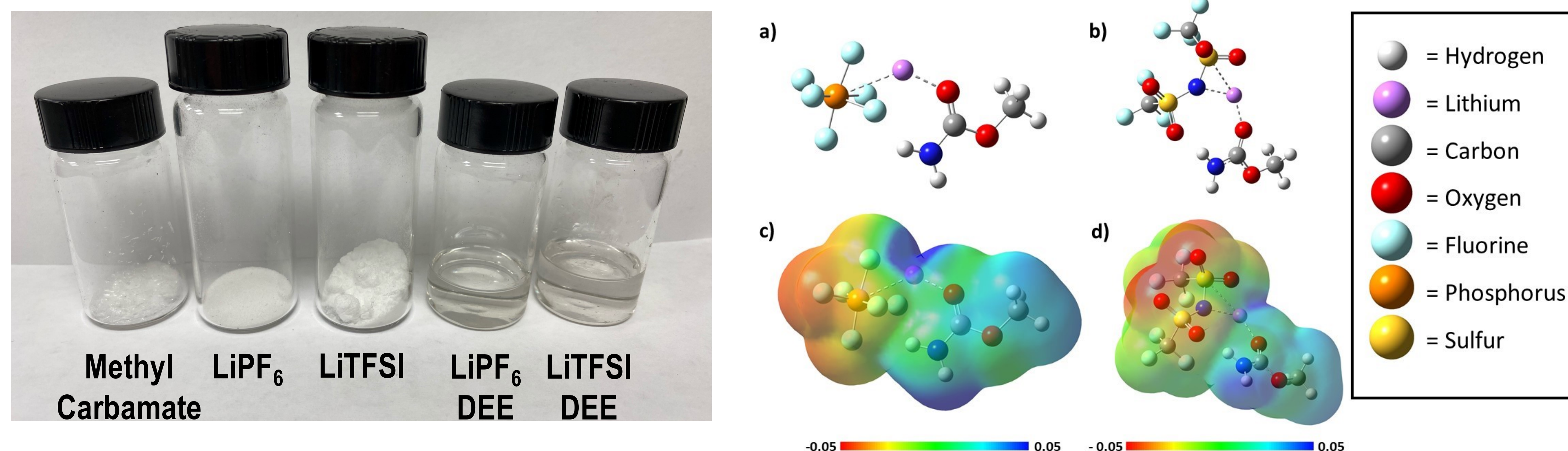


Fig 2: Left) Picture of chemicals used to make DEEs. From left to right: Methyl Carbamate Powder, Lithium hexafluorophosphate salt, Lithium bis(trifluoromethanesulfonyl)imide salt, LiPF₆ liquid DEE form after mixing, LiTFSI DEE formed after mixing.

Right) a) Coordination environment between LiPF₆ and methyl carbamate, b) coordination environment between LiTFSI and methyl carbamate, c) molecular electrostatic potential map of LiPF₆:methyl carbamate, and d) molecular electrostatic potential map of LiTFSI: methyl carbamate.

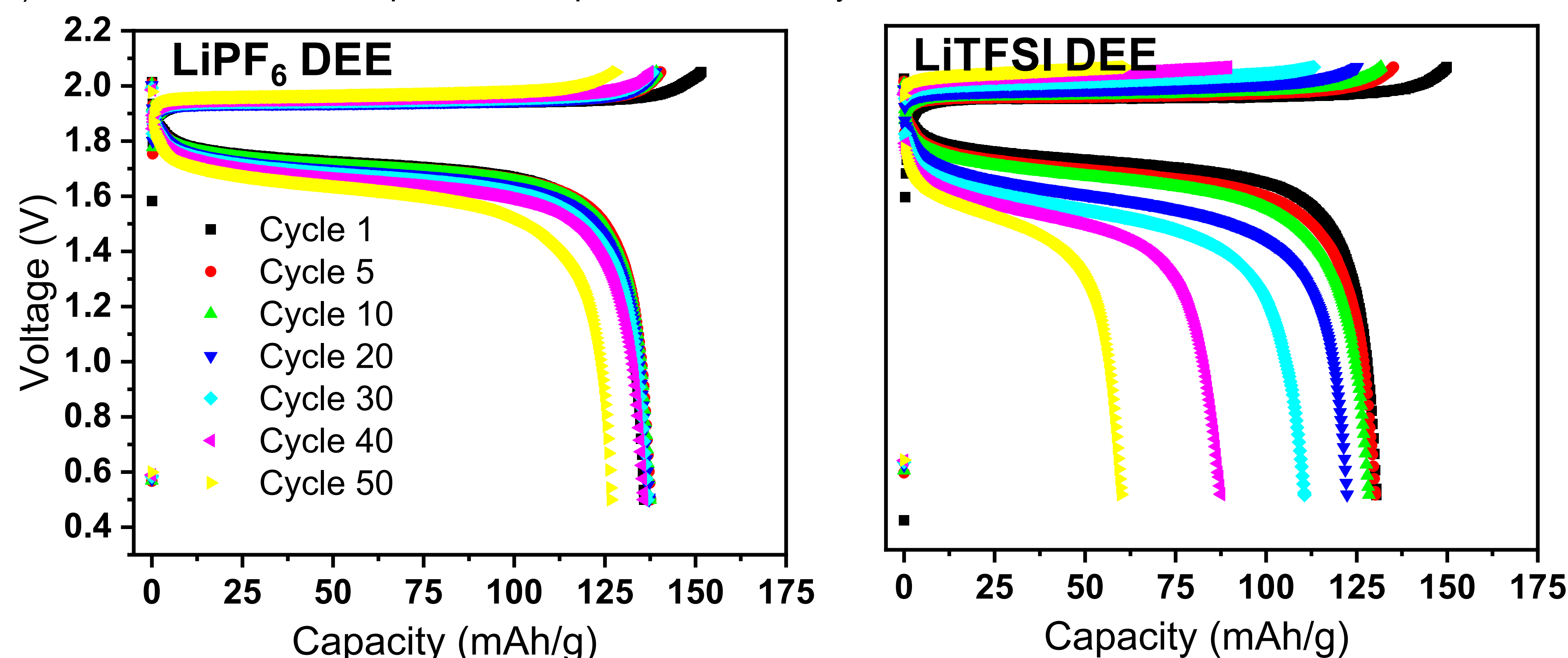


Fig 3: Cycling stability of formed DEEs using lithium titanate anode and lithium iron phosphate cathode. LiPF₆ DEE shows good cycling stability up to 50 cycles at 0.5C and LiTFSI showing degradation of cycling stability most likely due to less reductive stability (Figure 4)

Introduction

In the field of energy storage, electrolytes are the materials or liquids used to transport ions or redox active chemicals to facilitate an electrochemical process. While conventional electrolytes are generally inexpensive and work well with lithium-ion batteries, they have many shortfalls however, such as corrosion of the cell, flammability, toxicity, price, and incompatibility with other metals. Our work highlights the formation of a novel deep eutectic electrolyte (DEE) based on methyl carbamate and lithium salts. This DEE shows good cycling performance with the potential for low temperature operation. This study provides insights into a relatively unexplored field of non-conventional electrolyte systems.

Methods

The electrolytes, flooded cells, and coin cells were fabricated in an argon glovebox. We focused on the using commercial cell electrodes and used LiFePO₄ cathodes and LTO or Li metal anodes for cell fabrication. To test the electrochemical stability and performance of the DEE we used CV and constant current charging/discharging. To prepare the novel eutectic electrolyte based on methyl carbamate and lithium salts the starting materials were mixed at a 1:5 molar ratio (Li(PF₆ or TFSI) : Methyl carbamate at room temperature

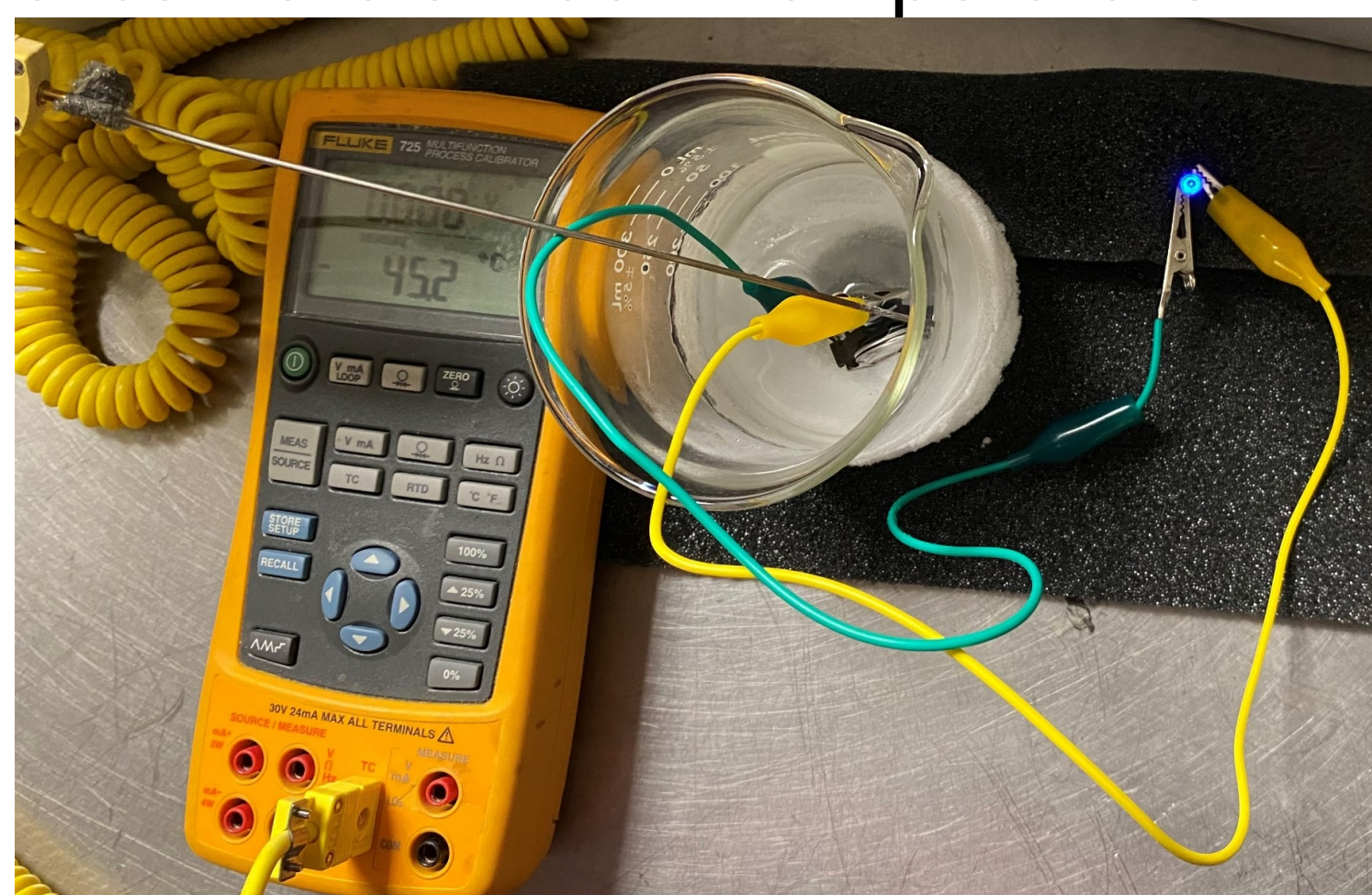


Fig 1: Low temperature coin cell operation

Acknowledgement

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Results

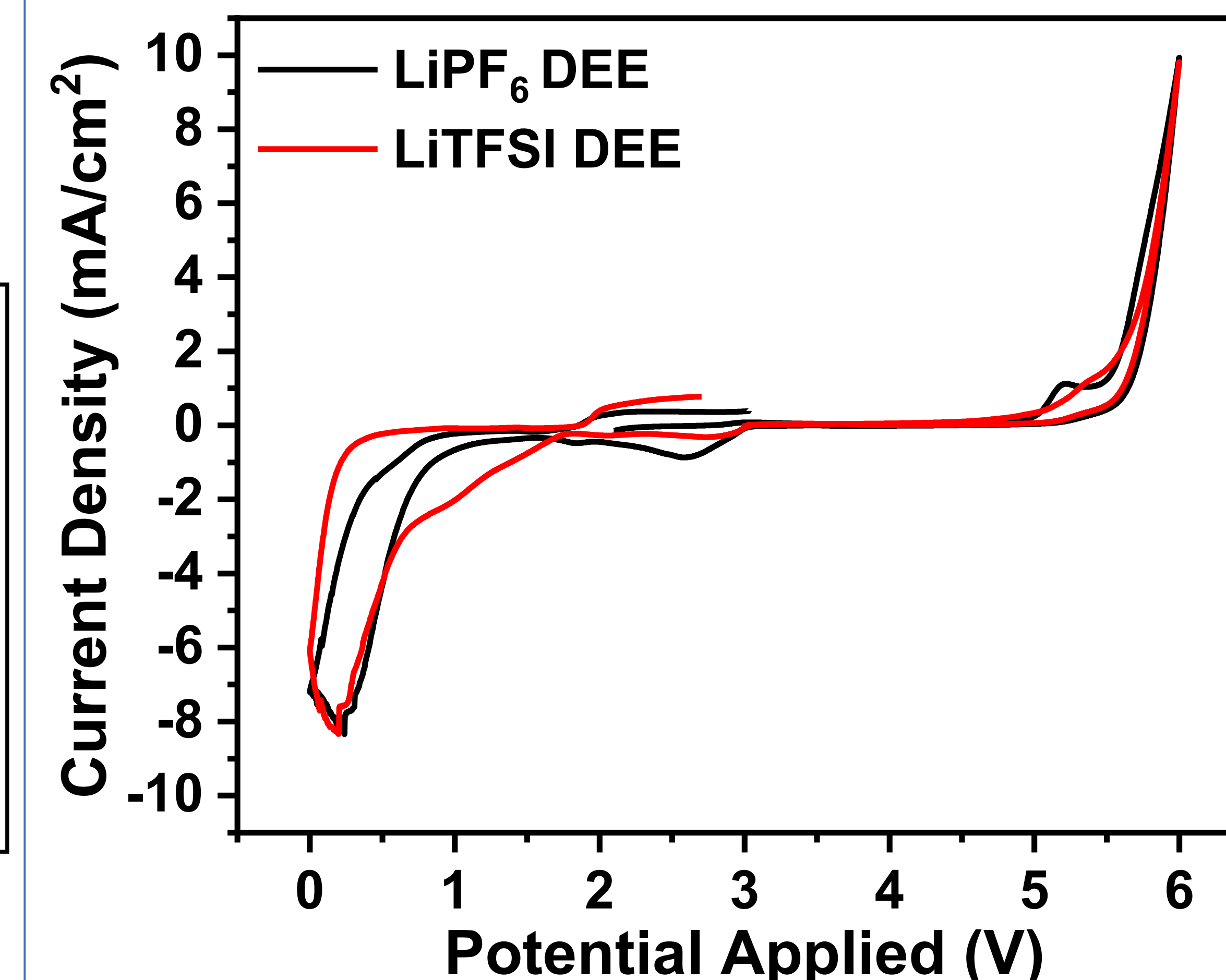


Fig 4: Pt|Li|Li three electrode flooded cell showing good oxidative stability of ~5V (versus Li/Li⁺). LiPF₆ DEE shows ~600 mV better reductive stability compared to LiTFSI DEE

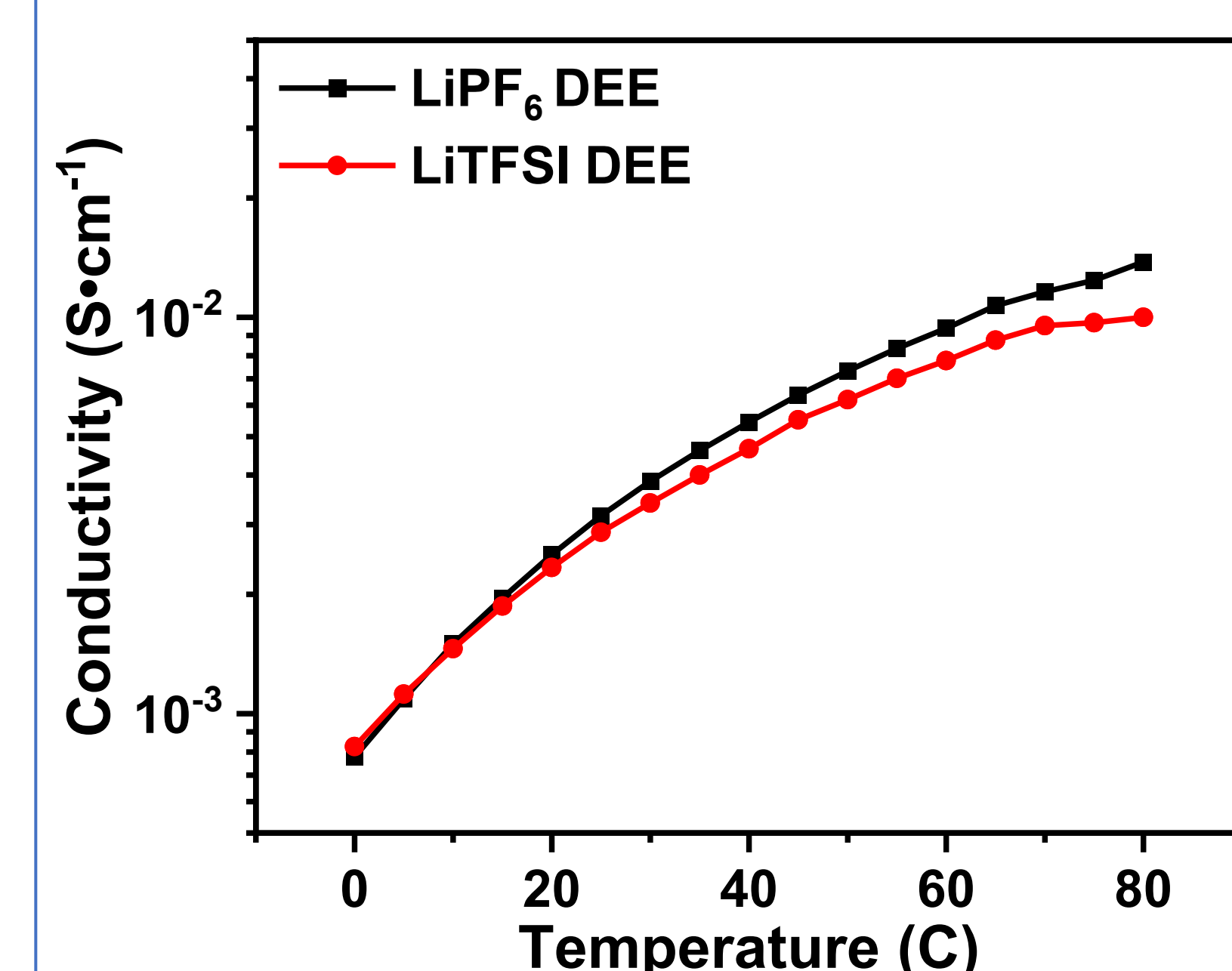


Fig 5: Conductivity at different temperatures of prepared DEEs showing very little difference between the two DEE systems

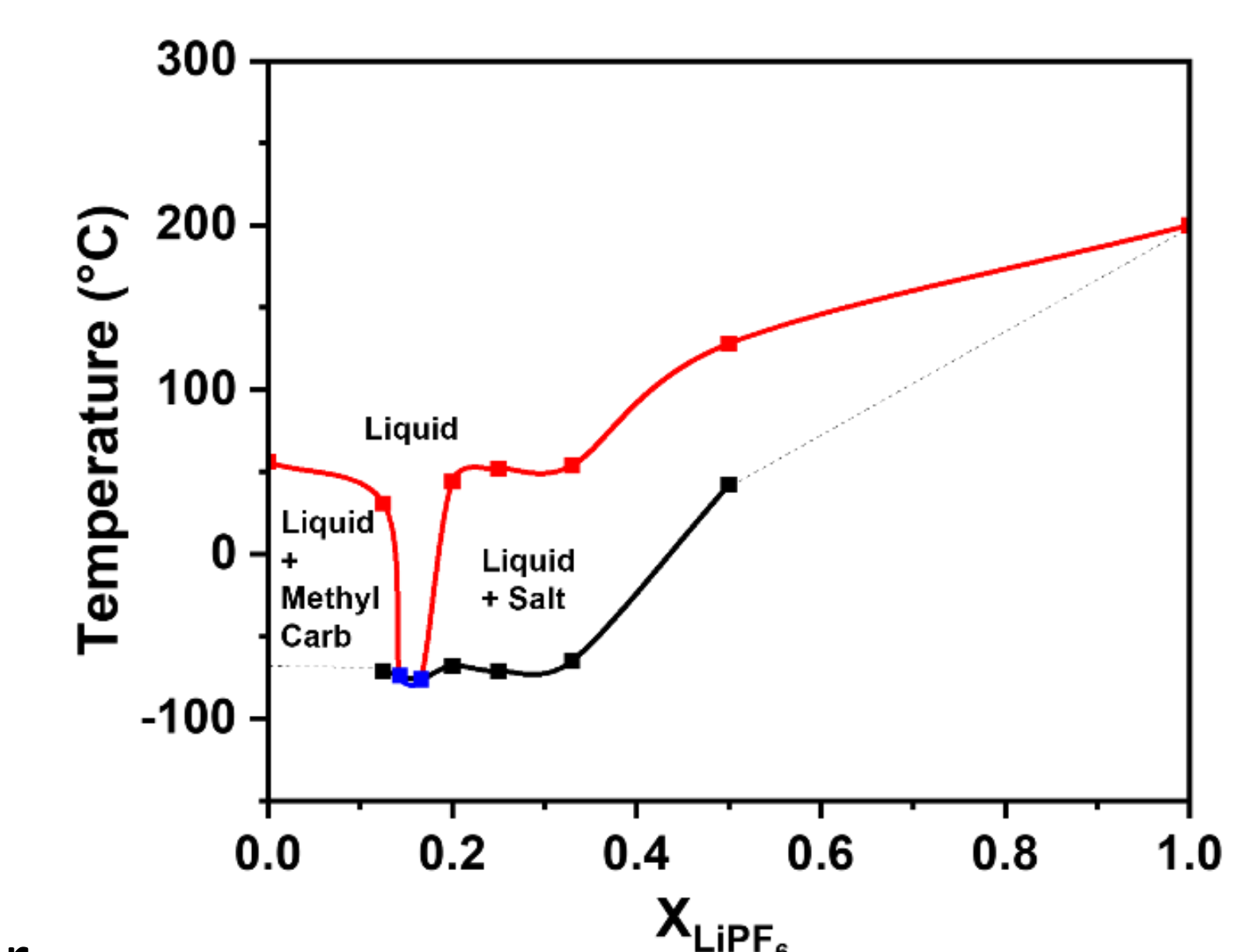


Fig 6: Phase Diagram of LiPF₆ DEEs at different ratios of salt to methyl carbamate