Improvement of Mechanical Strength of Solid-State Battery via Polymer Solid Electrolyte Composite

Abstract
Solid-state batteries (SSB) are a newly emerging interest in battery research for their improvement of conventional lithium-ion batteries with liquid electrolyte. They promise a replacement of volatile and flammable liquid electrolyte as well as greater energy storage due to the removal of a separator. Multiple aspects of solid-state batteries must still be tackled: low ion conductivity of the electrolyte, low mechanical strength, and most importantly, the issue of forming a stable electrode-electrolyte interface. Unlike liquid electrolytes, SSB’s solid electrolyte is more brittle and may not form a good interface with the electrode, which would make it harder for ion conduction. In order to address these issues, polymer solid electrolyte composite was examined – varying polymer addition methods, polymer compositions, and battery test parameters. It was important to find a balance of mechanical strength and conductivity.

Introduction
The key improvement of SSB from conventional battery with liquid electrolyte comes from the performance of solid electrolyte. A balance had to be found between mechanical strength and conductivity. The argyrodite (LiPSiCl), LPSCl) has effective conductivity by itself but it is brittle and lacks mechanical strength. Polymer on the other hand has zero to very small conductivity but is more elastic. Therefore, these properties were mixed and solid electrolyte composite was chosen, by adding polymer to LPSCl. Various methods of creating solid electrolyte were explored and to compare, the conductivity and mechanical strength were measured. The conductivity was measured for comparison using Swagelok cell configuration (Figure 1). Stainless Steel (SS) acted as a current collector. Their mechanical strength was also tested through cycling and indentation. In addition, the composition of the polymer could be varied when measuring conductivity and mechanical strength. The plot in Figure 2 shows correspondence in this concept. Increasing composition of polymer generally shows a decrease in overall conductivity, regardless of different composite processing methods.

Data and Results
After choosing the polymer composite and the processing method, the experiment continued into creating full coin cells with the LPSCl-polymer composite to mimic the real application of the solid state batteries. The baseline was established by testing with just the argyrodeite. Then during tests, different parameters were changed – increasing the pressure on the cell (by increasing the number of spacers in the coin cell) to improve the electrode-electrolyte interface, holding the cell voltage at certain values to maximize the capacity, and discharging faster. The use of coin cells for cycling allowed more tests to be conducted in a more effective manner.

Anticipated aspects of successful cycling result of full cells were high capacity (signaling high conductivity and energy storage), high efficiency (similar charging and discharging capacity), and more cycle numbers (without short circuiting which would show mechanical strength). Figure 3 plots the charge/discharge capacity across the cycles and the comparison between solid electrolyte composite with and without polymer. It was observed that the cell without polymer has higher overall capacity, as expected since polymer typically reduces the capacity of the batteries. Both cells show a great efficiency – negligible discrepancy between charging and discharging capacities.

Conclusions
There are several key takeaways gained from this research. First of all, thermal annealing improves conductivity. Comparing among all the tested methods, an overall increase in conductivity along with an increase in temperature was repeatedly noticed. This signals that conductivity can be improved even further when a polymer with thermal stability (no degradation at high temperature) is added. Secondly, uniform addition of polymer to LPSCl is crucial in determining the optimal method for solid electrolyte.

Addition of polymer is clearly essential in increasing mechanical strength but it simultaneously brings down the conductivity. When incorporating the solid electrolyte composite into an actual full cell test, the electrode-electrolyte interface plays a significant role in conduction and could be improved by increasing pressure on the cell.

Future additional research can be done to answer some lingering questions. The biggest concern of this research is reproducibility. There were many variations in the experiment and repeatability must be ensured before progressing onwards. In addition, exploration of more optimal method of polymer addition can be studied.

Acknowledgements
I would like to acknowledge Dr. Gao Liu for overseeing the projects as well as Dr. Piyachai Khoneim and Dr. Chen Fang for guiding the project and providing opportunities for skill developments.

References
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