

ABSTRACT - POSTER

Development of a self-healing solid composite electrolyte for lithium-ion batteries

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Lithium-solid-state batteries with high energy density play a key role in the energy transition. They are essential for storing renewable energy and for powering electric vehicles. Solid-state electrolytes offer significant advantages over liquid electrolytes, including enhanced thermal and electrochemical stability as well as improved safety. However, they often face challenges such as poor electrode wetting, dendrite formation, and physical cracks, which can be effectively addressed by using a self-healing electrolyte.

In this study, a silica-based solid electrolyte was developed, confining a LiTFSI-based ionic liquid electrolyte within a silica-based matrix while incorporating self-healing functionality. Self-healing was achieved by introducing an organic bridging agent, containing disulfide bonds, which can undergo reversible bond exchange. Covalent bonding between the organic and inorganic fractions of the solid electrolyte resulted in a stable, conductive network. The solid-liquid interface in the electrolytes was probed using Raman spectroscopy, which showed that the Li⁺-TFSI⁻ interaction was weakened due to their interaction with the solid matrix. The ionic conductivity of the electrolytes reached values up to 1.5 mS cm⁻¹, which is a high amount among self-healing electrolytes at room temperature (ex. 0.124 mS cm⁻¹). This improvement arises from using an inorganic silica matrix rather than a completely polymer-based system without a confined ionic liquid. The electrochemical stability window of these electrolytes was 1-5 V vs. Li⁺/Li.

To validate the self-healing properties, a testing method simulating in-battery scenarios was developed, demonstrating that the electrolytes exhibit efficient self-healing behavior. SEM images of deliberately damaged samples, taken before and after heating at 60 °C, also confirm that the material recovers its microstructure upon healing. Because the self-healing mechanism restores mechanical integrity, the organic fraction of the matrix can be reduced, allowing a higher silica content and a higher ionic conductivity. In addition, the gel is non-flammable, further enhancing the electrolyte's safety profile.

The self-healing electrolyte was incorporated into LFP/Li cells, which exhibited good rate capability (140 mAh g⁻¹ at 0.05C and 138 mAh g⁻¹ at 0.1C), and a long-term cycling stability test showed that the cell retained a high fraction of its capacity after 200 cycles. Stripping-plating experiments further demonstrate that the electrolyte remains stable against lithium metal in symmetric cells for more than 1000 cycles. Li Diffusion coefficient measurements on both the gel impregnated in a glass fiber separator and a pellet yield values around 9.5 x 10⁻⁸ cm²/s. Taken together, these results highlight the promise of this self-healing, silica-based solid electrolyte as a practical candidate for next-generation lithium batteries.

References

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