

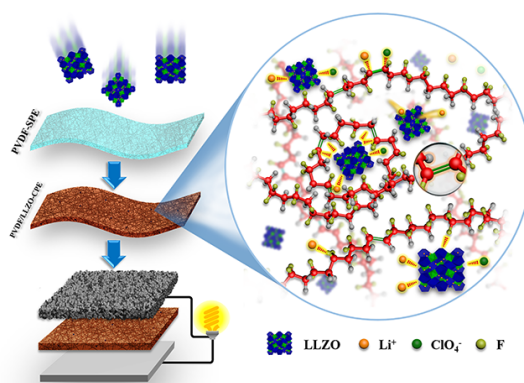
# Synergistic Coupling Between $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ and Poly(vinylidene fluoride) Induces High Electrochemical Performance of Solid Composite Electrolytes

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Easy processing and flexibility of polymer electrolytes make them very promising in developing all-solid-state lithium batteries. However, their low room-temperature conductivity, and poor mechanical and thermal properties still hinder their applications. Here, we use  $\text{Li}_{6.75}\text{La}_3\text{Zr}_{1.75}\text{Ta}_{0.25}\text{O}_{12}$  (LLZTO) ceramics to trigger structural modification of poly(vinylidene fluoride) (PVDF) polymer electrolyte. By combining experiments and first-principle calculations, we find that La atom of LLZTO could complex with N atom and C=O group of solvent molecule such as N,N-dimethylformamide along with electrons enriching at the N atom, which behaves like Lewis-base and induces the chemical dehydrofluorination of PVDF skeleton. Partially modified PVDF chains activate the interactions between PVDF matrix, lithium salt, and LLZTO fillers, hence leading to significantly improved performance of the flexible electrolyte membrane, e.g., a high ionic conductivity of about  $5 \times 10^{-4} \text{ S cm}^{-1}$  at  $25^\circ\text{C}$ , high mechanical strength, and good thermal stability. For further illustration, a solid-state lithium battery of  $\text{LiCoO}_2|\text{PVDF-based membrane}|\text{Li}$  is fabricated, and delivers satisfactory rate capability and cycling stability at room temperature. Our study indicates that the LLZTO modifying PVDF membrane is a promising electrolyte used for all-solid-state lithium batteries.



## References:

[1] X. Zhang, T. Liu, S. Zhang, X. Huang, B.Q. Xu, Y. Lin, B. Xu, L. Li, C.W. Nan, Y. Shen, *J. Am. Chem. Soc.* 139 (2017) 13779-13785.