Ionic Liquid Screening for Li-S Batteries

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Li-ion batteries are state-of-the-art energy storage systems widely used in commercial applications such as electric vehicles. However, these battery systems, by nature, cannot fulfill the energy demand in the future. Hence, post-Li-ion batteries, specifically lithium-sulfur (Li-S) batteries, are under investigation. Compared to the 800 Wh/kg specific energy of Li-ion batteries, Li-S batteries possess 2600 Wh/kg specific energy, which is more than three times the former. However, the practical energy densities obtained in Li-S batteries are far from their theoretical values, mainly due to low sulfur utilization and sulfur loss on the cathode side. The polysulfide (PS) shuttle mechanism (PSM), the parasitic shuttling of polysulfides from the cathode to the anode side, is a critical problem of Li-S batteries, resulting in sulfur escaping from the cathode and hence leads to irreversible capacity loss and low Coulombic efficiency. In this respect, preventing PSM is essential to improve the Li-S battery performance [1].

PSM effect is more severe when the concentration of polysulfides is high in the electrolyte; at this condition diffusion rate of PSs will be higher. Because of that, polysulfide concentration should be restricted to decrease the shuttling rate. Many alternatives exist to the most widely used organic electrolytes with high PS solubilities. Among these alternatives, ionic liquids (IL) have gained significant attention recently due to their high chemical and thermal stabilities and tailorable properties [1]. There are thousands of IL structures that depend on the selection of cation-anion pairs, which greatly affect PS solubilities and ionic liquid properties. While low PS solubility of ILs may be advantageous, most ionic liquids have high viscosities, hence low ionic transport. In this respect, selecting an optimum IL is a vital step. Since it is not possible to experimentally test all these ILs, COnductor like Screening MOdel for Real Solvents (COSMO-RS) calculations using COSMOThermX are performed for screening ILs regarding both PS solubilities and IL properties to be used as the liquid electrolytes of Li-S batteries in this study. 98 anions and 370 cations are readily available in the datasets of COSMOThermX, but many others are possible. Hence, using machine learning (ML), the relations between solubility-ionic liquid structural descriptors calculated from density functional theory (DFT) are also investigated to help design ionic liquid electrolytes.

There are 10 structural descriptors of each anion and cation present in the database. Association rule mining results, a commonly used ML method, show the associations between low PS solubility and the levels of each descriptor based on the lift values; high lift values signify stronger associations. As seen in Figure 1, while the level values get higher, higher lift values are obtained. Hence, cations with these descriptors should be utilized in Li-S batteries.

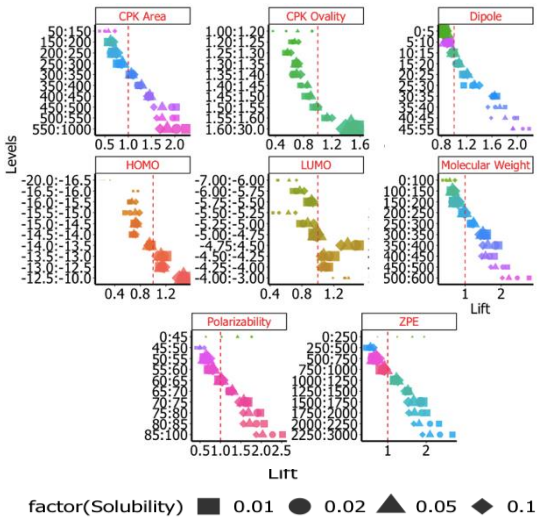

Figure 1. Levels of cation descriptors vs. lift values obtained from association rule mining analysis

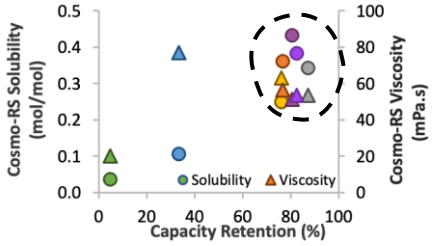

Figure 2. Relation between experimentally measured capacity retention of Li-S batteries vs. COSMO-RS predicted solubility and viscosity values.

This study shows the importance of the selection of cation-anion pairs of ILs for low PS solubility and viscosities. As seen in Figure 2, COSMO-RS calculations can be used to assess the final capacity retention of Li-S batteries.

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References
[1] Angulakshmi, N. et al. Frontiers in Energy Research, 3, (2015)

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