**Development of Recyclable Hybrid Solid Polymer Electrolytes**

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With efforts to decrease the impact of human activity on the environment leading to a drive to harness renewable energy sources, batteries have become an increasingly important means of storing energy prior to its use. From portable electronics to electric vehicles, and even providing power reserves to secure energy supplies to isolated cities like Adelaide, the impact of improvements in battery technology is clear. The vast majority of these batteries are lithium ion (Li-ion) batteries, which have dominated the field since their development in the 1970s-80s,1 recognized with the award of the Nobel Prize in Chemistry 2019 to Goodenough, Whittingham and Yoshino. Conventional Li-ion batteries use lithium salts in organic solvents, which pose some significant safety risks including fire and explosion. Replacing these flammable solvents with solid or semi-crystalline alternatives has been shown to improve the safety of Li-ion batteries via the development of all-solid-state batteries (ASSBs). Polymers that contain polar functional groups, such as ethers or carbonyl groups can function effectively as electrolytes by dissolving the metal ions, and facilitating their transport through the matrix, whilst displaying robust mechanical properties needed for use in everyday devices. Poly(ethylene oxide)s (PEO), for example, display considerable mechanical flexibility and chemical stability that makes them excellent candidate materials. However, a significant drawback to their widespread application and commercialization is their inability to meet the practical conductivities required (~10−3 S cm−1) due to frustrated transport of ions through the material. Effective transport of ions is thought to occur only in amorphous regions of the polymer, above the glass transition temperature, limiting the range of materials that can be used.2 Combining polymer electrolytes with ceramic fillers such as Al2O3 or TiO2 can drastically improve ionic conductivity without affecting mechanical strength. These hybrid polymer-inorganic electrolytes can possess outstanding mechanical properties, high ionic conductivities and excellent chemical and thermal stability, all of which are essential for next generation all-solid-state batteries.

Whilst battery technology has enabled the widespread adoption of renewable energy, batteries themselves present environmental challenges, particularly in their disposal, with only 2-3% of Li-ion batteries being recycled.3 The vast majority of batteries are underpinned by PEO, a high-performing, readily available commodity polymer. Recently, a method4 to enable the controlled cationic ring-opening polymerisation (CROP) of cyclic acetals such as 1,3-dioxolane has emerged as a route to polyethers that can be depolymerised upon treatment with acid, presenting a convenient route to their recycling. 1,3-dioxolane monomers can be sourced from biomass, presenting a sustainable route to the synthesis of recyclable polymer electrolytes.

The polyethers produced via CROP of 1,3-dioxolanes display structural similarities to PEO, and have already been demonstrated5 to function as polymer electrolytes in Li-ion batteries. In this project, we will explore the potential of poly(acetals) in hybrid electrolytes, assessing their ion transport mechanisms using a combination of conventional and *in situ* solid-state NMR spectroscopy, in conjunction with impedance measurements and muon spin relaxation spectroscopy studies. The effects of structural parameters of the polymers such as monomer composition and degree of polymerization on the resultant mechanical properties will be assessed, to enable the production of robust electrolytes. We will also evaluate a range of different inorganic ceramics to determine the optimal poly(acetal):ceramic combination. The performance of the poly(acetals) hybrid electrolytes prepared will be evaluated relative to current PEO-based electrolytes to determine their standing within the community of solid electrolytes.

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**Figure 1:** Controlled cationic ring-opening polymerization (CROP) of cyclic acetals can yield chemically recyclable polymers for use in hybrid electrolytes for Li-ion batteries.

**References**

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